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Title: **JP2002151150A2: POLYMER GEL ELECTROLYTE AND LITHIUM BA USING IT**

Country: JP Japan

Kind: A2 Document Laid open to Public inspection

Inventor: **RI YOHAN;**

Assignee: **SAMSUNG SDI CO LTD**
News, Profiles, Stocks and More about this company

Published / Filed: **2002-05-24** / 2001-09-05

Application Number: **JP2001000269134**

IPC Code: **H01M 10/40;** C08F 299/08; H01B 1/06; H01B 1/12; H01M 2/16; H01M 6/18;

Priority Number: 2000-09-05 **KR2000000052365**

Abstract:

PROBLEM TO BE SOLVED: To provide a polymer gel electrolyte capable of effectively inhibiting inflation caused by an electrolytic solution, and a lithium battery of high reliability and security.

SOLUTION: The polymer gel electrolyte is obtained by hardening of a composition containing either a polysiloxane compound or a polysiloxane- polyoxyalkylene compound, a polyethylene glycol derivative, and an organic solvent containing a lithium salt. The lithium battery used the electrolyte. The lithium battery is high in reliability and security because of its property by avoiding leaking of the electrolytic solution by effectively inhibiting inflation caused by the electrolytic solution.

INPADOC Legal Status: None **Get Now:** Family Legal Status Report

Family: Show 5 known family members

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(書誌＋要約＋請求の範囲)

(19)【発行国】日本国特許庁(JP)
(12)【公報種別】公開特許公報(A)
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(43)【公開日】平成14年5月24日(2002. 5. 24)
(54)【発明の名称】高分子ゲル電解質およびそれを用いてなるリチウム電池
(51)【国際特許分類第7版】

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H01M	2/16
	6/18

【FI】

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C08F	299/08	
H01B	1/06	A
	1/12	Z
H01M	2/16	L
		P
	6/18	E

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(33)【優先権主張国】韓国(KR)
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5G301 CA30 CD01 CE10
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5H029 AJ00 AJ15 AK03 AL06 AL07 AM00 AM02 AM03 AM05 AM07 AM16 BJ03 BJ14 BJ15 CJ11 EJ12 EJ14 HJ01

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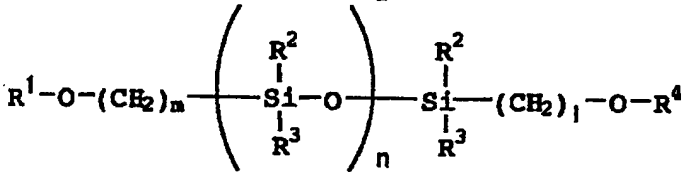
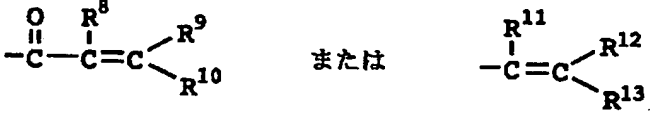
(57)【要約】
【課題】電解液による膨張現象を効果的に抑制しうる高分子ゲル電解質、および信頼性、安全性の高いリチウム電池を提供する。
【解決手段】ポリシロキサン化合物またはポリシロキサンーポリオキシアルキレン化合物と、ポリエチレングリコール誘導体と、リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなることを特徴とする高分子ゲル電解質、およびこれを用いたリチウム電池である。当該リチウム電池は、電解液による膨張現象を効果的に抑制するため、電解液が外部へ漏出し難いことを特徴としており、高い信頼性および安全性を有するものである。

【特許請求の範囲】
【請求項1】化学式1:【化1】

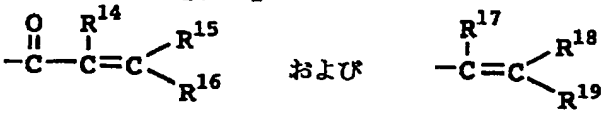


化学式1

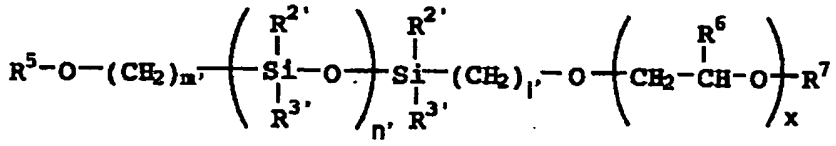
[式中、R^1は【化2】



であり、R^2およびR^3はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択され、R^4は、アリル、炭素数1～5の分岐を有していてもよいアルキル、【化3】

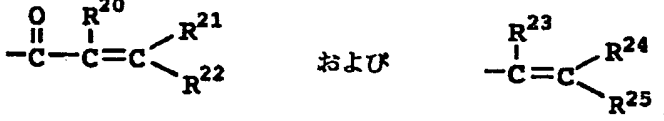


からなる群より選択される1つであり、R^8～R^{19}は、それぞれ独立して、水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、mは1～5であり、nは1～20であり、lは1～20である。]で示されるポリシロキサン化合物または化学式2:【化4】

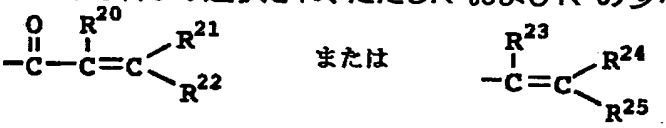


化学式2

[式中、$R^{2'}$および$R^{3'}$はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択され、R^5およびR^7はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、【化5】



からなる群より選択され、ただしR^5およびR^7の少なくとも1つは【化6】



であり、R^6は水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、m'は1～5であり、n'は1～20であり、l'は1～20であり、xは1～15であり、R^{20}～R^{25}は、それぞれ独立して、水素原子または炭素数1～5の分岐を有していてもよいアルキルである。]で示されるポリシロキサン－ポリオキシアルキレン化合物と、化学式3:【化7】

化学式3

[式中、R'は水素原子またはCH_3であり、R"は水素原子、－C(＝O)CH＝CH_2または－C(＝O)C(CH_3)＝CH_2であり、zは1～20である。]で示されるポリエチレングリコール誘導体と、リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなることを特徴とする高分子ゲル電解質。

【請求項2】前記ポリエチレングリコール誘導体は、ポリエチレングリコールジメタクリレート、ポリエチレングリコールジアクリレート、ポリエチレングリコールモノメタクリレートおよびポリエチレングリコールモノアクリレートからなる群より選択される一以上であることを特徴とする、請求項1に記載の高分子ゲル電解質。

【請求項3】前記化学式1で示されるポリシロキサン化合物または前記化学式2で示されるポリシロキサン－ポリオキシアルキレン化合物の含量が組成物100質量部を基準として0．1～10質量部であり、前記化学式3で示されるポリエチレングリコール誘導体の含量が0．4～50質量部であり、リチウム塩を含む有機溶媒の含量が50～97質量部であることを特徴とする、請求項1または2に記載の高分子ゲル電解質。

【請求項4】前記組成物は、エトキシ化トリメチロールプロパントリアクリレートをさらに含むことを特徴とする、請求項1～3のいずれか一項に記載の高分子ゲル電解質。

【請求項5】前記エトキシ化トリメチロールプロパントリアクリレートの含量が組成物100質量部を基準として0～5質量部であることを特徴とする、請求項4に記載の高分子ゲル電解質。

【請求項6】前記組成物は、ベンゾフェノン、過酸化ベンゾイル、過酸化アセチル、過酸化ラウロイル、およびアゾビスイソブチロニトリルからなる群より選択される一以上の重合開始剤を、組成物100質量部を基準として0．1～5質量部さらに含むことを特徴とする、請求項1～5のいずれか一項に記載の高分子ゲル電解質。

【請求項7】前記硬化は、熱重合、電子ビームによる重合、またはUVによる重合によってなされてなることを特徴とする、請求項1～6のいずれか一項に記載の高分子ゲル電解質。

【請求項8】前記熱重合において、重合温度が60～100℃であることを特徴とする、請求項7に記載の高分子ゲル電解質。

【請求項9】 前記リチウム塩は、$LiClO_4$、$LiBF_4$、$LiPF_6$、$LiAsF_6$、$LiCF_3SO_3$および$LiN(CF_3SO_2)_2$からなる群より選択される一以上であり、前記有機溶媒は、プロピレンカーボネート、エチレンカーボネート、ジメチルカーボネート、メチルエチルカーボネート、ジエチルカーボネート、ビニレンカーボネート、トリグリム、テトラグリムおよびγ－ブチロラクトンからなる群より選択される一以上であることを特徴とする、請求項1～8のいずれか一項に記載の高分子ゲル電解質。
【請求項10】 カソード、アノード、および、前記カソードと前記アノードとの間に介在するセパレータを含む電極組立体と、請求項1～9のいずれか一項に記載の高分子ゲル電解質と、前記電極組立体と前記高分子ゲル電解質とを内蔵するケースとを具備するリチウム電池。
【請求項11】 前記電極組立体は巻き取りにより形成されたものであり、前記ケースはパウチタイプであることを特徴とする、請求項10に記載のリチウム電池。
【請求項12】 前記セパレータは、ポリエチレンシート、ポリプロピレンシートまたはこれらを組み合わせたシートであることを特徴とする、請求項10または11に記載のリチウム電池。

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1/12		1/12	Z	5H024
H01M 2/16		H01M 2/16	L	5H029

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(22)出願日　平成13年9月5日(2001.9.5)
(31)優先権主張番号　00P52364
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(33)優先権主張国　韓国（KR）
(31)優先権主張番号　00P52365
(32)優先日　平成12年9月5日(2000.9.5)
(33)優先権主張国　韓国（KR）

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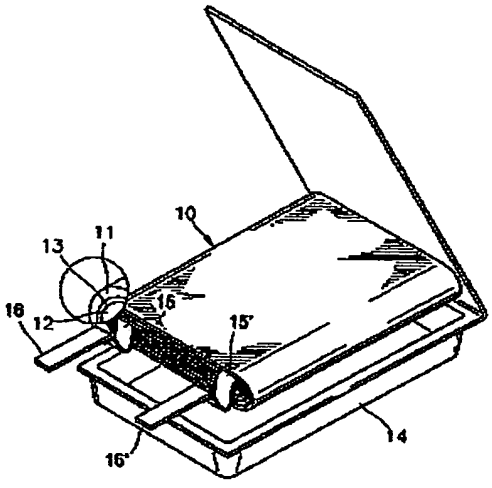
(74)代理人　100072349
弁理士　八田　幹雄　(外4名)

最終頁に続く

(54)【発明の名称】　高分子ゲル電解質およびそれを用いてなるリチウム電池

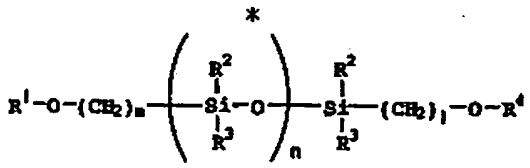
(57)【要約】
【課題】　電解液による膨張現象を効果的に抑制しうる高分子ゲル電解質、および信頼性、安全性の高いリチウム電池を提供する。
【解決手段】　ポリシロキサン化合物またはポリシロキサン－ポリオキシアルキレン化合物と、ポリエチレングリコール誘導体と、リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなることを特徴とする高分子ゲル電解質、およびこれを用いたリチウム電池である。当該リチウム電池は、電解液による膨張現象を効果的に抑制するため、電解液が外部へ漏出し難いことを特徴としており、高い信頼性および安全性を有するものである。



1 2

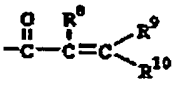
【特許請求の範囲】

【請求項1】 化学式1：

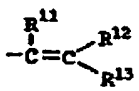


化学式1

[式中、R'は

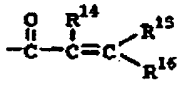


または

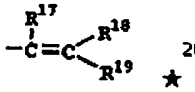


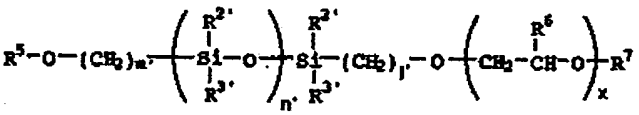
であり、R⁴およびR⁵はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択され、R⁶は、アリル、炭素数1～5の分岐を有していてもよいアルキル、

【化3】



および

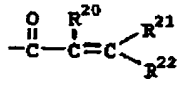




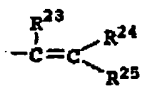
化学式2

[式中、R⁴'およびR⁵'はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択され、R⁵'および

【化5】



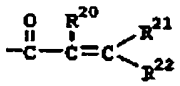
および



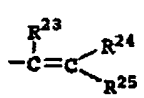
からなる群より選択され、ただしR⁵'およびR⁵'の少なくとも1つは

◆【化6】

◆



または



であり、R⁹は水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、m'は1～5であり、n'は1～20であり、l'は1～20であり、xは1～15であり、R¹⁰～R¹³は、それぞれ独立して、水素原子または炭素数1～5の分岐を有していてもよいアルキルである。]で示されるポリシロキサン－ポリオキシアルキレン化合物と、化学式3：

【化7】

★からなる群より選択される1つであり、R¹～R¹³は、それぞれ独立して、水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、mは1～5であり、nは1～20であり、lは1～20である。]で示されるポリシロキサン化合物または化学式2：

【化4】

☆R⁵'はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、

【化5】

$$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$$

化学式3

[式中、R'は水素原子またはCH₃であり、R''は水素原子、－C(=O)CH=CH₂または－C(=O)C(CH₃)=CH₂であり、zは1～20である。]で示されるポリエチレングリコール誘導体と、

リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなることを特徴とする高分子ゲル電解質。

【請求項2】 前記ポリエチレングリコール誘導体は、

ポリエチレングリコールジメタクリレート、ポリエチレングリコールジアクリレート、ポリエチレングリコールモノメタクリレートおよびポリエチレングリコールモノアクリレートからなる群より選択される一以上であることを特徴とする．請求項１に記載の高分子ゲル電解質。

【請求項３】 前記化学式１で示されるポリシロキサン化合物または前記化学式２で示されるポリシロキサン－ポリオキシアルキレン化合物の含量が組成物１００質量部を基準として０．１～１０質量部であり、前記化学式３で示されるポリエチレングリコール誘導体の含量が０．４～５０質量部であり．リチウム塩を含む有機溶媒の含量が５０～９７質量部であることを特徴とする、請求項１または２に記載の高分子ゲル電解質。

【請求項４】 前記組成物は、エトキシ化トリメチロールプロパントリアクリレートをさらに含むことを特徴とする．請求項１～３のいずれか一項に記載の高分子ゲル電解質。

【請求項５】 前記エトキシ化トリメチロールプロパントリアクリレートの含量が組成物１００質量部を基準として０～５質量部であることを特徴とする、請求項４に記載の高分子ゲル電解質。

【請求項６】 前記組成物は、ベンゾフェノン．過酸化ベンゾイル、過酸化アセチル、過酸化ラウロイル．およびアゾビスイソブチロニトリルからなる群より選択される一以上の重合開始剤を．組成物１００質量部を基準として０．１～５質量部さらに含むことを特徴とする、請求項１～５のいずれか一項に記載の高分子ゲル電解質。

【請求項７】 前記硬化は．熱重合．電子ビームによる重合．またはＵＶによる重合によってなされてなることを特徴とする．請求項１～６のいずれか一項に記載の高分子ゲル電解質。

【請求項８】 前記熱重合において．重合温度が６０～１００℃であることを特徴とする、請求項７に記載の高分子ゲル電解質。

【請求項９】 前記リチウム塩は、$LiClO_4$、$LiBF_4$、$LiPF_6$、$LiAsF_6$、$LiCF_3SO_3$およびLiN$(CF_3SO_2)_2$からなる群より選択される一以上であり、前記有機溶媒は、プロピレンカーボネート．エチレンカーボネート．ジメチルカーボネート、メチルエチルカーボネート．ジエチルカーボネート．ビニレンカーボネート、トリグリム、テトラグリムおよびγ－ブチロラクトンからなる群より選択される一以上であることを特徴とする、請求項１～８のいずれか一項に記載の高分子ゲル電解質。

【請求項１０】 カソード．アノード．および．前記カソードと前記アノードとの間に介在するセパレータを含む電極組立体と．

請求項１～９のいずれか一項に記載の高分子ゲル電解質と．

前記電極組立体と前記高分子ゲル電解質とを内蔵するケースとを具備するリチウム電池。

【請求項１１】 前記電極組立体は巻き取りにより形成されたものであり、前記ケースはパウチタイプであることを特徴とする．請求項１０に記載のリチウム電池。

【請求項１２】 前記セパレータは．ポリエチレンシート．ポリプロピレンシートまたはこれらを組み合わせたシートであることを特徴とする、請求項１０または１１に記載のリチウム電池。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】本発明は高分子ゲル電解質およびそれを用いてなるリチウム電池に関し、より詳細には．良好なゲル状を保ち得る高分子ゲル電解質を使用して安全性と信頼性とを向上させたリチウム電池に関する。

【０００２】

【従来の技術】リチウム電池は、従来のニッケルカドミウム電池およびニッケル水素原子電池に比べて．単位体積当たりのエネルギー密度および電圧が高く、電池の重さもそれらの半分程度であり、充放電サイクル寿命特性が優れており．かつ環境に悪影響を与えないことを特徴とする。そのためリチウム電池は次世代高性能バッテリとして関心を集めており．携帯用電子機器の小型軽量化および長時間使用への貢献が期待されている。

【０００３】リチウム電池は、液体電解質を使用するリチウムイオン電池と、ポリマー状の電解質を使用するリチウムイオンポリマー電池とに大別できる。リチウムイオン電池は、例えば円筒形または角形のケース内に電極組立体を密封するものである。しかし最近では．このようなケースを用いる代わりにパウチによって電極組立体を密封する方法が注目されている。パウチの使用により．単位質量および体積当たりのエネルギー密度をさらに高めることができるので．電池の薄型化および軽量化が可能であり．材料費も削減できる利点がある。

【０００４】図１は、一般的なパウチを使用したリチウムイオン電池の一例を模式的に示した分解斜視図である。図１で示されるリチウムイオン電池は、カソード１１．アノード１２およびセパレータ１３を含む電極組立体１０と、この電極組立体１０を包んで密封するケース１４とから成る。この場合．電極組立体１０は．カソード１１とアノード１２との間にセパレータ１３を挿入したものを巻き取ることによって形成されている。さらに．電極組立体１０と外部との電気的通路の役割を果たすカソードタップ１５およびアノードタップ１５'は、カソード１１およびアノード１２から引き出され．電極端子１６，１６'を形成する。

【０００５】図２は、従来のリチウムイオンポリマー電池の一例を模式的に示した分解斜視図である。図２で示されるリチウムイオンポリマー電池は．カソード．アノードおよびセパレータを含む電極組立体２１と．電極組

立体２１を包んで密封するケース２２とから成る．この場合．電極組立体２１で生じた電流を外部へ誘導するための電気的通路の役割を果たす電極端子（またはリード線）２４，２４´が、カソードおよびアノードに備えられたカソードタップ２３およびアノードタップ２３´に連結されており．これらはケース２２の外へ所定長さに延長される。

【０００６】このような図１のリチウムイオン電池および図２のリチウムイオンポリマー電池は、電極端子１６．１６´または２４，２４´の一部分のみを外部に露出させたまま．ケース１４または２２内に電極組立体１０または２１を入れて、ここに電解液を注入した後、熱と圧力とを加えて上部ケースの縁部と下部ケースの縁部とを熱接着性物質により接着させて密封することにより製造される。

【０００７】ここで沸点が低い有機溶媒を含む電解質を使用すると、電極組立体やパウチが膨張する現象が発生し．電池の信頼性および安全性が低下する、という問題がある。

【０００８】このような問題を解決するために．平面形電池を紫外線（ＵＶ）や電子ビームで硬化させて作るか．または、電解液を別途に注入しない方法、すなわちゲルを電極管に予めコーティングする方法等が提案されている（米国特許第５，９７２，５３９号、米国特許第５，２７９，９１０号、米国特許第５，４３７，９４２号および米国特許第５，３４０，３６８号）。しかしこれらの方法でも．膨張現象を満足すべき水準で防止、緩和することはできなかった。

【０００９】

【発明が解決しようとする課題】本発明の目的は．電解液による膨張現象を効果的に抑制しうる高分子ゲル電解質を提供することである．また本発明の他の目的は、このような高分子ゲル電解質を用いて．信頼性、安全性の高いリチウム電池を提供することである。

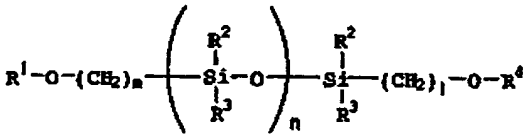
【００１０】

【課題を解決するための手段】本発明者らは、ゲル状電解質を調製するために従来用いられてきたポリエチレンオキシド、ポリプロピレンオキシドを主鎖とし．アクリル．ビニル、エポキシ基等の官能基を有するポリマーの代わりに、シロキサン単位またはオキシアルキレン単位を含有するポリシロキサン化合物またはポリシロキサン－ポリオキシアルキレン化合物を含む組成物を硬化することにより、物理的特性および電気化学的特性に優れると共に、膨張を効果的に抑制することのできる電解質が得られることを見出し、本発明を完成するに到った。

【００１１】従って本発明は、化学式１：

【００１２】

【化８】

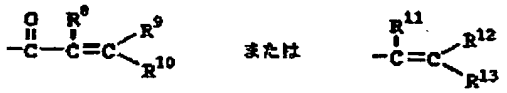


化学式１

【００１３】［式中、R¹は

【００１４】

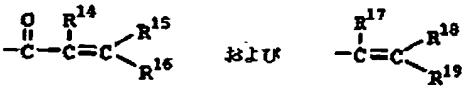
※【化９】



【００１５】であり、R³およびR⁴はそれぞれ独立して炭素数１～５の分岐を有していてもよいアルキル．フェニル．ベンジルおよびアリルからなる群より選択され、R⁵は、アリル、炭素数１～５の分岐を有していてもよいアルキル、

【００１６】

【化１０】

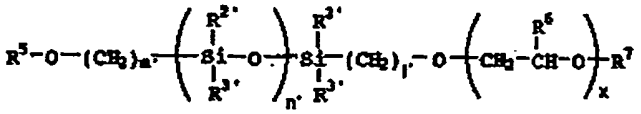


【００１７】からなる群より選択される１つであり、R⁶～R¹⁹は．それぞれ独立して．水素原子または炭素数１～５の分岐を有していてもよいアルキルであり．mは１～５であり．nは１～２０であり．lは１～２０である。］で示されるポリシロキサン化合物、または化学式２：

【００１８】

【化１１】

7 8

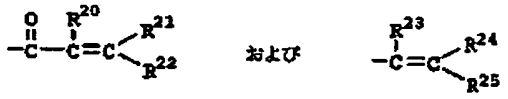


化学式2

【0019】〔式中、R²'およびR³'はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択され．R'およびR'はそれぞれ独立して炭素数1～5の分＊岐を有していてもよいアルキル、

【0020】

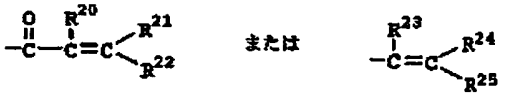
【化12】



【0021】からなる群より選択され．ただしR'およびR'の少なくとも1つは

※【0022】

※【化13】



【0023】であり、R'は水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、m'は1～5であり、n'は1～20であり．l'は1～20であり．xは1～15であり．R²⁰～R²⁵は、それぞれ独立して、水素原子または炭素数1～5の分岐を有していてもよいアルキルである。〕で示されるポリシロキサン－ポリオキシアルキレン化合物と、化学式3：

【0024】

【化14】

$$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$$

化学式3

【0025】〔式中、R'は水素原子またはCH₃であり．R''は水素原子、－C（＝O）CH＝CH₂または－C（＝O）C（CH₃）＝CH₂であり、zは1～20である。〕で示されるポリエチレングリコール誘導体と．リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなることを特徴とする高分子ゲル電解質である。

【0026】さらに本発明は、前記ポリエチレングリコール誘導体は．ポリエチレングリコールジメタクリレート．ポリエチレングリコールジアクリレート、ポリエチレングリコールモノメタクリレートおよびポリエチレングリコールモノアクリレートからなる群より選択される一以上であることを特徴とする、前記高分子ゲル電解質である。

【0027】さらに本発明は、前記化学式1で示されるポリシロキサン化合物または前記化学式2で示されるポリシロキサン－ポリオキシアルキレン化合物の含量が組成物100質量部を基準として0．1～10質量部であり．前記化学式3で示されるポリエチレングリコール誘導体の含量が0．4～50質量部であり、リチウム塩を含む有機溶媒の含量が50～97質量部であることを特徴とする、前記高分子ゲル電解質である。

【0028】さらに本発明は、前記組成物は、エトキシ化トリメチロールプロパントリアクリレートをさらに含むことを特徴とする、前記高分子ゲル電解質である。

【0029】さらに本発明は、前記エトキシ化トリメチロールプロパントリアクリレートの含量が組成物100質量部を基準として0～5質量部であることを特徴とする．前記高分子ゲル電解質である。

【0030】さらに本発明は、前記組成物は、ベンゾフェノン、過酸化ベンゾイル．過酸化アセチル、過酸化ラウロイル、およびアゾビスイソブチロニトリルからなる群より選択される一以上の重合開始剤を、組成物100質量部を基準として0．1～5質量部さらに含むことを特徴とする、前記高分子ゲル電解質である。

【0031】さらに本発明は、前記硬化は、熱重合、電子ビームによる重合、またはUVによる重合によってなされてなることを特徴とする、前記高分子ゲル電解質である。

【0032】さらに本発明は、前記熱重合において、重合温度が60～100℃であることを特徴とする．前記高分子ゲル電解質である。

【0033】さらに本発明は、前記リチウム塩は．LiClO₄．LiBF₄、LiPF₆．LiAsF₆．LiCF₃SO₃およびLiN（CF₃SO₂）₂からなる群より選択される一以上であり．前記有機溶媒は、プロピレンカーボネート．エチレンカーボネート．ジメチルカーボネート、メチルエチルカーボネート．ジエチルカーボネ

ート．ビニレンカーボネート、トリグリム、テトラグリムおよびγ－ブチロラクトンからなる群より選択される一以上であることを特徴とする、前記高分子ゲル電解質である。

【００３４】さらに本発明は、カソード、アノード、および．前記カソードと前記アノードとの間に介在するセパレータを含む電極組立体と、前記高分子ゲル電解質と．前記電極組立体と前記高分子ゲル電解質とを内蔵するケースとを具備するリチウム電池である。

【００３５】さらに本発明は、前記電極組立体は巻き取りにより形成されたものであり、前記ケースはパウチタイプであることを特徴とする、前記リチウム電池である。

【００３６】さらに本発明は、前記セパレータは．ポリエチレンシート．ポリプロピレンシートまたはこれらを＊

＊組み合わせたシートであることを特徴とする、前記リチウム電池である。

【００３７】

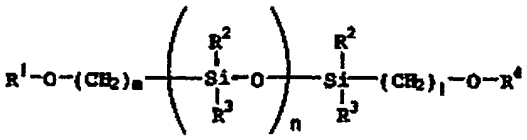
【発明の実施の形態】本発明の高分子ゲル電解質は、ポリシロキサン化合物、ポリシロキサン－ポリオキシアルキレン化合物と．ポリエチレングリコール誘導体と、リチウム塩を含有する有機溶媒とを含む組成物を硬化させてなる。当該組成物は、以下、「高分子ゲル電解質を形成するための組成物」または単に「組成物」とも称する。

【００３８】まず組成物に含まれる各成分について説明する。

【００３９】ポリシロキサン化合物は．化学式１：

【００４０】

【化１５】

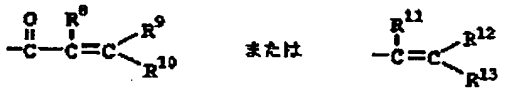


化学式１

【００４１】で示される。式中、R¹は

【００４２】

※【化１６】



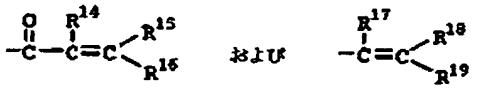
【００４３】であり、R⁸～R¹¹は水素原子または炭素数１～５の分岐を有していてもよいアルキルである。R⁸として具体的には．－Ｃ（＝Ｏ）ＣＨ＝ＣＨ₂．－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂、－Ｃ（＝Ｏ）ＣＨ＝ＣＨＣＨ₃が挙げられ、好ましくは－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂である。

【００４４】R⁹およびR¹⁰は、それぞれ独立して．炭素数１～５の分岐を有していてもよいアルキル、フェニル．ベンジルおよびアリルからなる群より選択される。R⁹およびR¹⁰として具体的には、メチル、エチル．プロピル．イソプロピル、ｎ－ブチル、ｓｅｃ－ブチル、ｔｅｒ－ブチル．ペンチル．イソペンチルが挙げられ、好ましくはメチル．エチル．フェニルまたはベンジルである。

【００４５】R¹¹は．アリル（－ＣＨ₂ＣＨ＝ＣＨ₂）．炭素数１～５の分岐を有していてもよいアルキル．

【００４６】

【化１７】



【００４７】からなる群より選択される１つであり、R¹²～R¹³は水素原子または炭素数１～５の分岐を有していてもよいアルキルである。R¹²として具体的には、－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂、－Ｃ（＝Ｏ）ＣＨ＝ＣＨＣＨ₃．メチル、エチル、プロピル、イソプロピル．ｎ－ブチル、ｓｅｃ－ブチル、ｔｅｒ－ブチル．ペンチル．イソペンチルが挙げられ、好ましくはメチル．－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂である。

【００４８】mは１～５である。nは１～２０．好ましくは１～１０である。lは１～２０．好ましくは１～１０である。m．nおよびlがこの範囲である時．化学式１で示される化合物は、溶解度、反応性および電池性能面で優れているため好ましい。

【００４９】このようなポリシロキサン化合物として、R¹が－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂、R²がメチル、R³がメチル、R⁴がメチル．m、n．およびlが３であるポリシロキサン化合物．R¹が－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂．R²がエチル、R³がメチル、R⁴がメチル．mが２または３、nが３または５．lが３または５であるポリシロキサン化合物、R¹が－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂、R²がフェニル、R³がメチル．R⁴がメチル、mが２または３．nおよびlが３または５であるポリシロキサン化合物．R¹が－Ｃ（＝Ｏ）Ｃ（ＣＨ₃）＝ＣＨ₂．R²がベンジル．R³がメチル、R⁴がメチル．mが２または３、nおよびlが３または５である

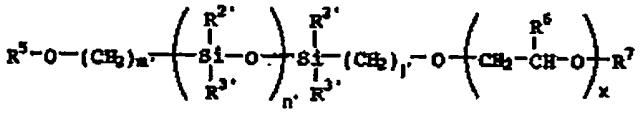
ポリシロキサン化合物、R¹が-C(=O)C(CH₃)=CH₂、R²がフェニル、R³がメチル、R⁴がエチル、m=2または3、n=3または5、l=3または5であるポリシロキサン化合物、R¹が-C(=O)C(CH₃)=CH₂、R²がベンジル、R³がメチル、R⁴がエチル、mが2または3、nおよびlが3または5であるポリシロキサン化合物、が溶解度、反応性および電池性能面で優れているため好ましく用いられ、これらのなか*

*でも特にR¹が-C(=O)C(CH₃)=CH₂、R²がメチル、R³がメチル、R⁴がメチル、m、nおよびlが3であるポリシロキサン化合物が好ましい。

【0050】次にポリシロキサン-ポリオキシアルキレン化合物について説明する。当該化合物は、化学式2：

【0051】

【化18】



化学式2

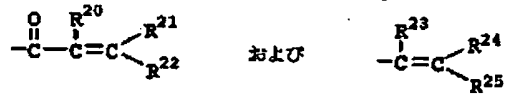
【0052】で示される。式中、R²'およびR³'はそれぞれ独立して炭素数1～5の分岐を有していてもよいアルキル、フェニル、ベンジルおよびアリルからなる群より選択される。具体的には、メチル、エチル、プロピル、イソプロピル、n-ブチル、sec-ブチル、ter-ブチル、ペンチル、イソペンチルなどが挙げられ、※

※好ましくはメチル、エチル、フェニルまたはベンジルである。

【0053】R¹およびR⁴は、それぞれ独立して、炭素数1～5の分岐を有していてもよいアルキル、

【0054】

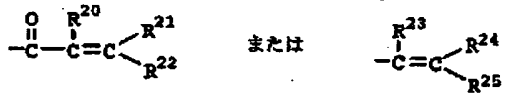
【化19】



【0055】からなる群より選択され、ただしR¹およびR⁴の少なくとも1つは、

★【0056】

★【化20】



【0057】であり、R²⁰～R²⁵はそれぞれ独立して水素原子または炭素数1～5の分岐を有していてもよいアルキルである。R¹またはR⁴として具体的には、-C(=O)C(CH₂)=CH₂、-C(=O)CH=CHCH₃、メチル、エチル、プロピル、イソプロピル、n-ブチル、sec-ブチル、ter-ブチル、ペンチル、イソペンチルなどが挙げられ、好ましくは-C(=O)C(CH₃)=CH₂またはメチルである。

【0058】R⁵は水素原子または炭素数1～5の分岐を有していてもよいアルキルであり、該アルキルとして具体的には、メチル、エチル、プロピル、イソプロピル、n-ブチル、sec-ブチル、ter-ブチル、ペンチル、イソペンチルなどが挙げられる。R⁵として好ましくは水素原子である。

【0059】m'は1～5である。n'は1～20であり、好ましくは1～10である。l'は1～20であり、好ましくは1～10である。xは1～15である。m'、n'、l'およびxがこの範囲である時、化学式2で示される化合物は、溶解度、反応性および電池性能面で優れているため好ましい。

【0060】このような化学式2で示されるポリシロキサン-ポリオキシアルキレン化合物として、R²'がメチル、R³'がメチル、R¹が-C(=O)C(CH₂)=CH₂、R⁵が水素原子、R₇がメチル、m'、n'および l'が3、xが5であるポリシロキサン-ポリオキシアルキレン化合物、R²'がエチル、R³'がメチル、R¹が-C(=O)C(CH₂)=CH₂、R⁵が水素原子、R₇がメチル、m'が2または3、n'および l'がそれぞれ3または5、xが3、5または10であるポリシロキサン-ポリオキシアルキレン化合物、R²'がベンジル、R³'がメチル、R¹が-C(=O)C(CH₃)=CH₂、R⁵が水素原子、R⁷がメチル、m'が2または3、n'および l'がそれぞれ3または5、xが3、5または10であるポリシロキサン-ポリオキシアルキレン化合物、R²'がフェニル、R³'がメチル、R¹が-C(=O)C(CH₂)=CH₂、R⁵が水素原子、R⁷がメチル、m'が2または3、n'および l'がそれぞれ3または5、xが3、5または10であるポリシロキサン-ポリオキシアルキレン化合物、R²'がベンジル、R³'がメチル、R¹が-C(=O)C(CH₂)=CH₂、R⁵

が水素原子、R''がメチル、m'が2または3、n'および1'がそれぞれ3または5、xが3、5または10であるポリシロキサン－ポリオキシアルキレン化合物が溶解度、反応性および電池性能面で優れているため好ましく用いられる。これらのなかでも特にR''がメチル、R''がメチル、R'が－C（＝O）C（CH_2）＝CH_2、R''が水素原子、R_7がメチル、m'、n'および1'が3、xが5であるポリシロキサン化合物が好ましい。

【0061】化学式1で示されるポリシロキサン化合物または化学式2で示されるポリシロキサン－ポリオキシアルキレン化合物の含量は、組成物100質量部を基準として0．1～10質量部であることが好ましい。ここで含量が0．1質量部未満の場合、これら化合物の付加による電池の性能改善の効果が低く、10質量部を超える場合、電池の性能が低下する恐れがある。

【0062】化学式1で示されるポリシロキサン化合物は、例えば、ジヒドロキシ末端ポリシロキサンとアクリロイル塩化物とを反応させて、ポリシロキサン化合物を製造することができるが、これに限定されず、当業界周知の方法で製造することができる。同様に、化学式2で示されるポリシロキサン－ポリオキシアルキレン化合物を様々な方法により製造可能である。例えば、ジヒドロキシ末端ポリシロキサンとエチレンオキシドとを反応させて製造されるポリシロキサン－ポリオキシアルキレンにアクリロイル塩化物を反応させて、ポリシロキサン－ポリオキシアルキレン化合物を製造することができるが、これに限定されず、当業界周知の方法で製造することができる。

【0063】次にポリエチレングリコール誘導体について説明する。当該ポリエチレングリコール誘導体は、化学式3：

【0064】

【化21】

$$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$$

化学式3

【0065】で示される。式中、R'は水素原子またはCH_3であり、R''は水素原子、－C（＝O）CH＝CH_2または－C（＝O）C（CH_3）＝CH_2であり、zは1～20である。このようなポリエチレングリコール誘導体として好ましくは、ポリエチレングリコールジメタクリレート、ポリエチレングリコールジアクリレート、ポリエチレングリコールモノメタクリレートおよびポリエチレングリコールモノアクリレートなどが挙げられる。これらは、それぞれ単独で用いてもよいし、混合して用いてもよい。特にポリエチレングリコールジメタクリレート、ポリエチレングリコールモノメタクリレート、または、それらの混合物を用いることによって、電池の性能を高めることができるので好ましい。

【0066】当該ポリエチレングリコール誘導体の含量の範囲は、組成物100質量部を基準として0．4～50質量部が好ましい。ここで含量が0．4質量部未満の場合、硬化反応性が低下し、50質量部を超える場合、電池の性能が低下する恐れがある。しかしながらポリエチレングリコール誘導体の含量の範囲は用いるものによって様々なのでこの限りではない。以下、具体的に好ましい含量の範囲を述べる。例えば、ポリエチレングリコールジメタクリレートの含量は組成物100質量部を基準として0．5～50質量部であることが好ましく、ポリエチレングリコールモノメタクリレートの含量は組成物100質量部を基準として0．4～50質量部であることが好ましい。

【0067】次に本発明のリチウム塩を含有する有機溶媒について説明する。リチウム塩としては、過塩素酸リチウム（$LiClO_4$）、四フッ化ホウ素リチウム（$LiBF_4$）、六フッ化リンリチウム（$LiPF_6$）、六フッ化砒素リチウム（$LiAsF_6$）、三フッ化メタンスルホン酸リチウム（$LiCF_3SO_3$）およびリチウムビストリフルオロメタンスルホニルアミド（$LiN(CF_3SO_2)_2$）からなる群より選択される一以上であることが好ましい。特に好ましくは、$LiPF_6$である。もちろん上記以外にも当該技術分野で周知のリチウム塩が使用可能である。

【0068】有機溶媒としては、プロピレンカーボネート、エチレンカーボネート、ジメチルカーボネート、メチルエチルカーボネート、ジエチルカーボネート、ビニルレンカーボネート、トリグリム、テトラグリムおよびγ－ブチロラクトンからなる群より選択される一以上であることが好ましい。特に好ましくは、エチレンカーボネート（EC）、ジメチルカーボネート（DMC）、ジメチルカーボネート（DEC）であり、これらを単独で用いてもよいし、適切な体積比で混合して用いてもよい。もちろん上記以外にも当該技術分野で周知の有機溶媒が使用可能である。

【0069】このようなリチウム塩を含む有機溶媒の含有量は、組成物100質量部を基準として50～97質量部であることが好ましい。かつ、リチウム塩の含有量は、有機溶媒1Lに対して0.5～3molであることが好ましい。ここで有機溶媒およびリチウム塩の含量が前記範囲を外れる場合、電池の性能が低下する恐れがある。

【0070】さらに本発明の組成物は、エトキシ化トリメチロールプロパントリアクリレート（ethoxylated trimethylol propane triacrylate）を含むことが好ましい。このような化合物を含むことにより、組成物の硬化反応を促進させることができる。エトキシ化トリメチロールプロパントリアクリレートの含量は、組成物100質量部を基準として0～5質量部であることが好ましく、ここで5質量部を超える場合には電池の性能が低下

する恐れがある。

【００７１】以上述べた化学式１で示されるポリシロキサン化合物または化学式２で示されるポリシロキサン－ポリオキシアルキレン化合物、ポリエチレングリコール誘導体、および、リチウム塩を含有する有機溶媒を混合することによって、本発明の高分子ゲル電解質を形成するための組成物を得ることができる。

【００７２】さらに前記組成物は、ベンゾフェノン、過酸化ベンゾイル、過酸化アセチル、過酸化ラウロイル、およびアゾビスイソブチロニトリルからなる群より選択される一以上の重合開始剤を含むことが好ましく、特にアゾビスイソブチロニトリルが一般的に用いられる。ここで重合開始剤の含量は、一般的には組成物１００質量部を基準として０．１～５質量部であるが、種類によってそれぞれの適切な含量を選択すべきである。

【００７３】本発明の高分子ゲル電解質は、上述の成分を含む組成物を硬化することによって製造される。硬化は、好ましくは熱重合、電子ビームによる重合、または、UVによる重合が用いられる。以下に各重合方法について詳細に説明する。

【００７４】熱重合は、重合温度に調節されたオーブンで組成物を所定の重合時間で処理することによって行われる。ここで重合温度は６０～１００℃であることが好ましく、ここで６０℃未満の場合、重合が不十分になる恐れがあり、一方１００℃を超過すると膨張現象が生じる可能性がある。重合時間は、例えばケース内で重合させるのか、それともフィルムにキャスティングした状態で重合させるのかによって適宜調節することが必要であるため限定されないが、一般的には４時間程度である。

【００７５】電子ビームによる重合は、電子ビームを組成物に所定の時間照射することによってなされる。該電子ビームの出力は通常１．５ｅＶ以上であり、ここで１．５ｅＶ未満の場合、長い硬化時間が必要になる、または硬化が不十分になるなどの恐れがある。電子ビームを照射する時間は一般的には３０秒以上であることが好ましく、時間が３０秒未満の場合、重合が不十分になる恐れがある。このような電子ビームによる重合は、上述の熱重合に比べて反応時間を短縮でき、さらに重合開始剤を必要としないという利点を有する。

【００７６】UVによる重合は、UVを組成物に所定の時間照射することによってなされる。該UVの波長は一般的には２５０～３６５ｎｍであることが好ましく、ここでこの範囲を外れると重合が不十分になる恐れがある。UVを照射する時間は一般的には３０秒以上であることが好ましく、時間が３０秒未満の場合、重合が不十分になる恐れがある。このようなUVによる重合は、上述の熱重合に比べて反応時間を短縮できる、という利点を有する。

【００７７】次に、上述した組成物を用いて高分子ゲル電解質を製造する方法を説明する。まず各成分を上述した含有量の範囲内で準備する。これらの含量は、製造されるリチウム電池の電気化学的安定性、電池の性能等を考慮して最適に選択されるべきである。次に化学式１で示されるポリシロキサン化合物または化学式２で示されるポリシロキサン－ポリオキシアルキレン化合物、化学式３で示されるポリエチレングリコール誘導体、および、エトキシ化トリメチロールプロパントリアクリレートを混合する。この混合物に、必要に応じて上記の重合開始剤をさらに添加し、さらにリチウム塩を含有する有機溶媒を添加し、攪拌して均一な組成物を得る。得られた組成物は、上述した熱重合、電子ビームによる重合、または、UVによる重合などによって硬化される。

【００７８】次に、本発明の高分子ゲル電解質を用いたリチウム電池について説明する。本発明のリチウム電池は、カソード、アノード、および、前記カソードと前記アノードとの間に介在するセパレータを含む電極組立体と、上述した本発明の高分子ゲル電解質と、前記電極組立体と前記高分子ゲル電解質とを内蔵するケースとを具備するものである。

【００７９】まずカソードまたはアノードとして用いるカソード活物質組成物およびアノード活物質組成物について説明すると、カソードには、一般的には$LiCoO_2$などのリチウム含有酸化物からなるカソード活物質組成物が好ましく使用される。アノードには、一般的にはカーボン、グラファイトなどを含むアノード活物質組成物が好ましく使用され、特にカーボンとしてメソカーボンファイバーが一般的に好ましい。

【００８０】さらにこれら活物質組成物は、導電剤、結合剤、溶媒を適宜含んでいてもよい。導電剤としては、これらに限定されないが、カーボンブラック等が好ましく使用される。ここで導電剤の含量は、電極活物質（例えば$LiCoO_2$）１００質量部を基準として１～２０質量部であることが好ましく、ここで含量が２０質量部を超える場合には電極活物質層の導電性が低下し、１質量部未満の場合には電極活物質の含量が相対的に低くなるために好ましくない。

【００８１】結合剤としては、ビニリデンフルオライド－ヘキサフルオロプロピレンコポリマー（VdF／HFPコポリマー）、ポリビニリデンフルオライド、ポリアクリロニトリル、ポリメチルメタクリルレートまたはその混合物が好ましく使用され、特にポリビニリデンフルオライドが一般的に好ましい。結合剤の含量は、電極活物質１００質量部を基準として２～３０質量部であることが好ましい。ここで含量が前記範囲の時、電極集電体に対する電極活物質の結合力に優れる。

【００８２】溶媒としては、一般的にリチウム電池に使用されるものであればいずれも使用可能であり、例えばアセトン、Ｎ－メチルピロリドンが挙げられ、他の成分に併せて適宜選択される。

【００８３】セパレータは、当該分野において用いられ

るものであれば特に制限されないが．ポリエチレンシート．ポリプロピレンシートまたはこれらを組み合わせたシートであることが好ましい。特に．ポリエチレンセパレータ、ポリプロピレン／ポリエチレン／ポリプロピレン三層セパレータが巻き取りやすいという理由で好ましく用いられる。具体的には．朝日化学工業社製のポリエチレンセパレータが用いられる。

【００８４】このようなカソード、アノードおよびセパレータを含む前記電極組立体と前記高分子ゲル電解質とを内蔵するケースは、当業界で用いられるタイプであればいずれも使用可能だが．特にパウチタイプがゲル電解質の外部漏洩を良好に防ぐことができるので好ましい。

【００８５】次に、本発明のリチウム電池の製造方法を説明する。まず．上述のアノード活物質組成物またはカソード活物質組成物を用いて集電体上に電極活物質層を形成し、カソード電極板およびアノード電極板を作製する。ここで電極活物質層は．各活物質組成物を集電体上に直接コーティングする方法や、例えばドクターブレードによって各活物質組成物を別途に支持体上部にコーティングおよび乾燥した後．この支持体から剥離して得られた活物質組成物のフィルムを集電体上にラミネーションする方法によって形成される。このような支持体は、活物質層が支持できるものならいずれも使用でき．例えばマイラーフィルム（デュポン社製）などのポリエチレンテレフタルレートフィルムが好適である。

【００８６】次に得られたカソード電極板とアノード電極板との間にセパレータを挿入し、電極組立体を形成する。ここで当該組立体を．ジェリーロール方式を用いた巻き取りによって形成しても（図１）．または．バイセル構造に形成してもよい（図２）。このようにして得られた組立体を．ケース内に設置する。

【００８７】次に、本発明の高分子ゲル電解質を形成するための組成物をケース内に仕込み．硬化することによって本発明の高分子ゲル電解質を形成する。ひとつの方法として、本発明の高分子ゲル電解質を形成するための組成物をケース内に注入し．密封して得られたケースを．上述した電子ビームによる重合またはＵＶによる重合によって硬化する方法が挙げられる。このようにケース内で組成物を重合する場合、熱重合による効果が最も効率よく組成物を硬化させることができる。

【００８８】他の方法として、アノード電極板またはカソード電極板のどちらか一方、またはこれら両方の表面に．本発明の電解質を形成するための組成物を例えばドクターブレードによってキャスティングした後．上述した電子ビームによる重合またはＵＶによる重合によって硬化する方法も好ましい。このようにして、本発明のリチウム電池を得ることができる。

【００８９】本発明のリチウム電池は．リチウム１次電池．またはリチウムイオンポリマー電池またはリチウムイオン電池などのリチウム２次電池のいずれにも適用することができる。

【００９０】

【実施例】以下．実施例を挙げて本発明をより詳細に説明する。

【００９１】＜高分子ゲル電解質およびそれを用いたリチウム電池の作製＞

実施例１

アセトン６００ｍｌにポリビニリデンフルオライド１５ｇを添加して．ボールミルで２時間混合して溶解させて混合物を得た。この混合物に$LiCoO_2$４７０ｇと．アセチレンブラック（商品名：スーパーＰ、MMM社製）１５ｇとを添加し、５時間混合してカソード活物質組成物を作製した。

【００９２】前記カソード活物質組成物を、３２０μｍのギャップを有するドクターブレードを使用して厚さ１４７μｍ、幅４．９ｃｍのアルミニウム薄膜上にコーティングし、乾燥して、単位カソード電極板を作製した。

【００９３】一方、アセトン６００ｍｌにポリビニリデンフルオライド５０ｇを添加してボールミルで２時間混合し．溶解させ．混合物を得て、この混合物にメソカーボンファイバー４４９ｇとシュウ酸１ｇとを添加し、５時間混合してアノード活物質組成物を作製した。

【００９４】前記アノード活物質組成物を、４２０μｍのギャップを有するドクターブレードを使用して厚さ１７８μｍ、幅５．１ｃｍの銅薄膜上にコーティングし、乾燥して、単位アノード電極板を作製した。

【００９５】前記カソード電極板と前記アノード電極板との間に、幅５．２５ｃｍ．厚さ１８μｍのポリエチレンセパレータ（朝日化学工業社）を介在させ、これをジェリーロール方式で巻き取って電極組立体を作製した。この電極組立体をパウチに入れた。

【００９６】一方、化学式１においてR^1．R^2およびR^3がメチル．R^3が－Ｃ（＝Ｏ）Ｃ（CH_2）＝CH_2、ｍ．ｎおよび１が３であるポリシロキサン化合物０．２ｇ．ポリエチレングリコールジメタクリルレート１．８ｇ．ポリエチレングリコールモノメタクリルレート０．５ｇ．エトキシ化トリメチロールプロパントリアクリレート０．５ｇ．アゾビスイソブチロニトリル０．１ｇ、および、エチレンカーボネート（ＥＣ）／ジメチルカーボネート（ＤＭＣ）／ジメチルカーボネート（ＤＥＣ）＝３：３：１（体積比）であり$LiPF_6$を１Ｍ含む有機溶媒３０ｇを混合して．高分子ゲル電解質を形成するための組成物を調製した。この組成物３ｇを上述の方法により得られた電極組立体が入ったパウチに注入し、密封した。その結果物を８０℃に調節したオーブンで４時間処理することにより、本発明のリチウム電池を完成させた。

【００９７】実施例２

アセトン６００ｍｌにポリビニリデンフルオライド１５ｇを添加してボールミルで２時間混合し、溶解させ、混

合物を得た。この混合物に$LiCoO_2$ 470gとスーパーP（MMM社製）15gを添加した後、これを5時間混合してカソード活物質組成物を作製した。

【0098】得られたカソード活物質組成物を、320μmのギャップを有するドクターブレードを使用して、厚さ147μm、幅4.9cmのアルミニウム薄膜上にコーティングし、乾燥して単位カソード電極板を作製した。

【0099】その後、実施例1と同じポリシロキサン化合物0.2g、ポリエチレングリコールジメタクリルレート1.8g、ポリエチレングリコールモノメタクリルレート0.5g、エトキシ化トリメチロールプロパントリアクリレート0.5g、ベンゾフェノン0.1g、およびEC：DMC：DEC=3：3：1（体積比）でありLiPF$_6$を1Mで含む有機溶媒30gを混合して、高分子ゲル電解質を形成するための組成物を製造した。この組成物を上記で製造したカソード電極板にドクターブレードを用いてキャスティングした後、365nmのUVを1.5時間、照射して硬化させ、高分子ゲル電解質が形成されたカソード電極を得た。

【0100】一方、アセトン600mlにポリビニリデンフルオライド50gを添加してボールミルで2時間混合し、溶解させ、組成物を得た。この混合物にメソカーボンファイバー449gとシュウ酸1gとを添加し、5時間混合して、アノード活物質組成物を調製した。

【0101】前記アノード活物質組成物を420μmのギャップを有するドクターブレードを使用して、厚さ178μm、幅5.1cmの銅薄膜上にコーティングし、乾燥して、単位アノード電極板を作製した。

【0102】高分子ゲル電解質が形成された前記カソード電極板と前記アノード電極板との間に、幅5.25cm、厚さ18μmのポリエチレンセパレータ（朝日化学工業社）を介在させた後、これをジェリーロール方式で巻き取って電極組立体を作製した。この電極組立体をパウチ内に入れてリチウム電池を完成させた。

【0103】実施例3

高分子ゲル電解質を形成するための組成物を、カソード電極板の代わりにアノード電極板にキャスティングし、硬化させたことを除いては実施例2と同じ方法でリチウム電池を作製した。

【0104】実施例4

高分子ゲル電解質を形成するための組成物を、カソード電極板およびアノード電極板の両方にキャスティングしたことを除いては実施例2と同じ方法でリチウム電池を作製した。

【0105】実施例5

アセトン600mlにポリビニリデンフルオライド15gを添加してボールミルで2時間混合し、溶解させ、混合物を得た。この混合物に$LiCoO_2$ 470gとスーパーP（MMM社製）15gとを添加し、5時間混合して、カソード活物質組成物を調製した。

【0106】前記カソード活物質組成物を320μmのギャップを有するドクターブレードを使用して、厚さ147μm、幅4.9cmのアルミニウム薄膜上にコーティングし、乾燥して、単位カソード電極板を作製した。

【0107】一方、アセトン600mlにポリビニリデンフルオライド50gを添加してボールミルで2時間混合し、溶解させて混合物を得た。この混合物にメソカーボンファイバー（MCF）449gとシュウ酸1gとを添加し、5時間混合してアノード活物質組成物を調製した。

【0108】前記アノード活物質組成物を420μmのギャップを有するドクターブレードを使用して、厚さ178μm、幅5.1cmの銅薄膜上にコーティングし、乾燥して、単位アノード電極板を作製した。

【0109】前記カソード電極板と前記アノード電極板との間に厚さ18μm、幅5.25cmのポリエチレンセパレータ（朝日化学工業社）を介在させた後、これをゼルーロール方式で巻き取って電極組立体を作製した。この電極組立体をパウチ内に入れた。

【0110】一方、化学式2において$R^{2'}$、$R^{3'}$およびR_4がメチル、$R^{1'}$が$-C(=O)C(CH_2)=CH_2$、$R^{5'}$が水素原子、m'、n'およびl'が3、xが5のポリシロキサン-ポリオキシアルキレン化合物0.2g、ポリエチレングリコールジメタクリルレート1.8g、ポリエチレングリコールモノメタクリルレート1g、エトキシ化トリメチロールプロパントリアクリレート0.05g、アゾビスイソブチロニトリル0.01g、および、EC：DMC：DEC=3：3：1（体積比）でありLiPF$_6$を1Mで含む有機溶媒30gを混合して、高分子ゲル電解質を形成するための組成物を製造した。この組成物3gを、実施例1で得られたパウチ電池に注入し、これを密封した。続いて、結果物を80℃に調節したオーブンで4時間処理することによりリチウム電池を作製した。

【0111】実施例6

アセトン600mlにポリビニリデンフルオライド15gを添加してボールミルで2時間混合し、溶解させて混合物を得た。この混合物に$LiCoO_2$ 470gとスーパーP（MMM社製）15gとを添加し、5時間混合して、カソード活物質組成物を調製した。

【0112】前記カソード活物質組成物を、320μmのギャップを有するドクターブレードを使用して厚さ147μm、幅4.9cmのアルミニウム薄膜上にコーティングし、乾燥して、単位カソード電極板を作製した。

【0113】その後、実施例5と同一なポリシロキサン-ポリオキシアルキレン化合物0.2g、ポリエチレングリコールジメタクリルレート1.8g、ポリエチレングリコールモノメタクリルレート1g、エトキシ化トリメチロールプロパントリアクリレート0.05g、ベン

ゾフェノン0．01g、および、EC：DMC：DEC＝3：3：1（体積比）でありLiPF6を1Mで含む有機溶媒30gを混合して．高分子ゲル電解質を形成するための組成物を調製した。この組成物を、上記で製造したカソード電極板にドクターブレードを用いてキャスティングした後．波長365nmのUVを1．5時間照射して硬化させた。

【0114】一方、アセトン600mlにポリビニリデンフルオライド50gを添加してボールミルで2時間混合し．溶解させて混合物を得た。この混合物にメソカーボンファイバー（MCF）449gとシュウ酸1gとを添加し、5時間混合してアノード活物質組成物を調製した。

【0115】前記アノード活物質組成物を、420μmのギャップを有するドクターブレードを使用して．厚さ178μm、幅5．1cmの銅箔膜上にコーティングし．乾燥して．単位アノード電極板を作製した。

【0116】高分子ゲル電解質が形成されたカソード電極板とアノード電極板との間に、幅5．25cm．厚さ18μmのポリエチレンセパレータ（朝日化学工業社）を介在させた後．これをジェリーロール方式で巻き取って電極組立体を作製した。この電極組立体をパウチ内に入れてリチウム電池を作製した。

【0117】実施例7

高分子ゲル電解質を形成するための組成物をカソード電極板の代わりにアノード電極板にキャスティングしたことを除いては実施例6と同じ方法でリチウム電池を作製した。

【0118】実施例8

高分子ゲル電解質を形成するための組成物をカソード電極板とアノード電極板の両方にキャスティングしたことを除いては実施例6と同じ方法でリチウム電池を作製した。

【0119】比較例

本発明の高分子ゲル電解質を形成するための組成物の代わりに、1M LiPF6およびEC／DMC／DECを体積比3：3：4で含む混合溶液（宇部興産社製）を使用したことを除いては．実施例1と同じ方法によりリチウム電池を作製した。

【0120】＜電池の特性評価＞

試験方法

実施例1～8および比較例のリチウム電池は、寿命特性試験．貫通試験．高温放置（85℃）時の膨張試験、圧力40kgf／cm²下での漏液試験により．信頼性および安全性を評価した。

【0121】寿命特性試験は、実施例1～8および比較例1のリチウム電池を用いて充放電を100サイクル行い．それによる電池の体積変化を観察することによりなされた。この評価は、体積変化が少ないことを好ましいとした。

【0122】貫通実験は．0．2Cの電流で3時間充電後．電池の長軸に垂直な方向に直径5mmの釘をその中央に貫通させて電池の発火現象および破裂現象の有無を調べた。

【0123】膨張試験は．0．2Cの電流で3時間充電後．85℃で4時間放置した後、電池の厚さを測定して評価した。放置後の電池の厚さが初期厚さの110%以下ならば良好とした。

【0124】漏液試験は．電池を40kgf／cm²の圧力で10秒間加圧した後．漏液の有無を調べた。

【0125】試験結果

図3は、実施例1および比較例のリチウム電池の．寿命特性を示すグラフである。これによれば、実施例1の電池の体積減少は．比較例とほぼ同程度であった。また貫通試験においては、発火や破裂は起こらず、さらに膨張試験においても電池の初期厚さの110%を保っており．さらに漏液試験においても中から液漏れすることはなかった。実施例2～8もほぼ同様の結果を示したことから．本発明のリチウム電池は優れた寿命特性を有することがわかった。

【0126】以上より、実施例1～8のリチウム電池は．電解液が優れたゲル状を保つことができるために、電解液の外部漏洩や、電解液による電極組立体やパウチの膨張を抑制することができ、比較例の電池より優れた信頼性および安全性を有することがわかった。

【0127】

【発明の効果】本発明の高分子ゲル電解質は、シロキサン単位またはオキシアルキレン単位を含有するポリシロキサン化合物またはポリシロキサン－ポリオキシアルキレン化合物を含む組成物を硬化してなることにより、物理的特性および電気化学的特性に優れると共に．膨張を効果的に抑制することができるリチウム電池を提供し得るものである。当該電解質を用いて得られたリチウム電池は．電解質の外部漏洩や．電解液による電極組立体やパウチの膨張が抑制されるために、高い信頼性および安全性を有するものである。

【図面の簡単な説明】

【図1】　一般的なパウチを使用したリチウムイオン電池の一例を模式的に示した分解斜視図である。

【図2】　従来のリチウムイオンポリマー電池の一例を模式的に示した分解斜視図である。

【図3】　実施例1および比較例のリチウム電池の寿命特性を測定した結果を示すグラフである。

【符号の説明】

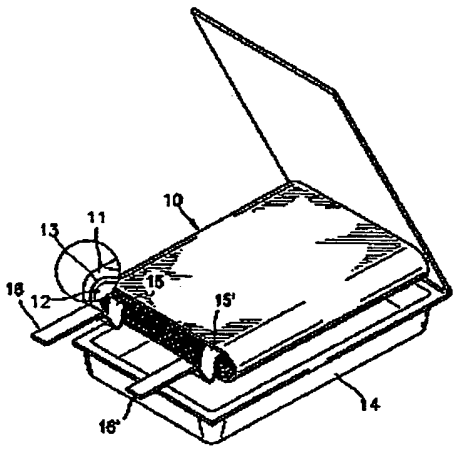
10、21	電極組立体
11	カソード
12	アノード
13	セパレータ
14、22	ケース
15、23	カソードタップ

23　　24

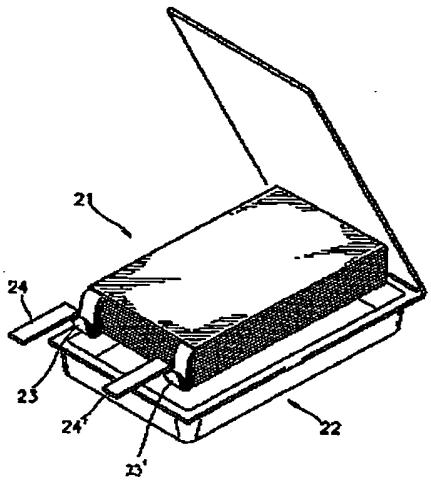
15', 23'　アノードタップ　　＊ード線)

16. 16'. 24. 24'　電極端子（またはリ＊

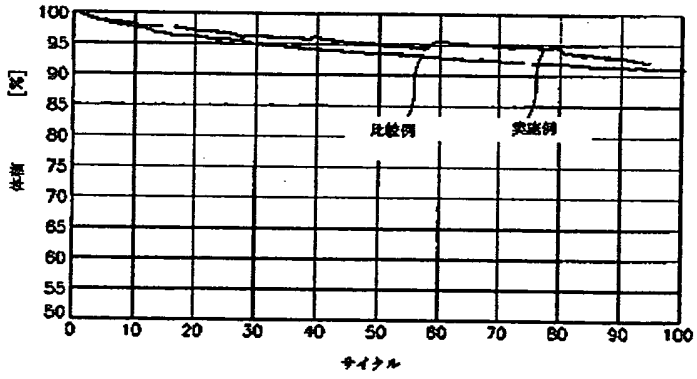
【図１】



【図２】



【図３】



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H01M 2/16		H01M 2/16	P	
6/18		6/18	E	

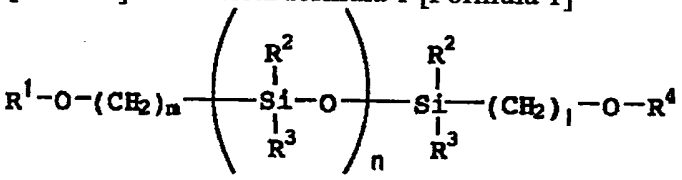
Ｆターム(参考) 4J027 AA04 AC03 AF05 BA26 CA12
CB10 CC02 CC03 CC05 CC06
CD00
5G301 CA30 CD01 CE10
5H021 AA06 CC04 EE04
5H024 AA00 AA02 BB11 CC04 CC12
CC13 DD09 EE09 FF14 FF15
FF18 FF19 FF23 FF36 HH01
5H029 AJ00 AJ15 AK03 AL06 AL07
AM00 AM02 AM03 AM05 AM07
AM16 BJ03 BJ14 BJ15 CJ11
EJ12 EJ14 HJ01

* NOTICES *

1. This document has been translated by computer.So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3.In the drawings, any words are not translated.

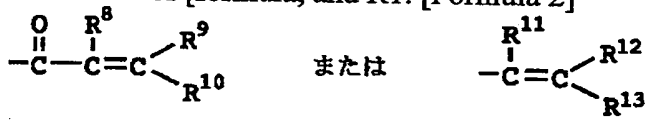
CLAIMS

[Claim(s)]
[Claim 1] : Chemical formula 1 [Formula 1]

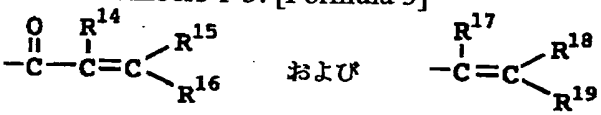


化学式1

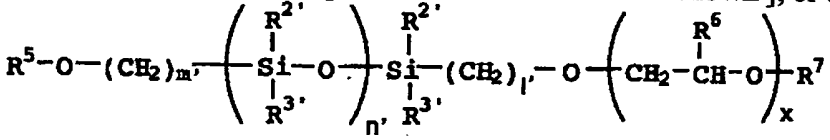
The inside of [formula, and R1. [Formula 2]



It is the alkyl in which it appears, and exists, R2 and R3 are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound, and R4 may have branching of an allyl compound and carbon numbers 1-5. [Formula 3]

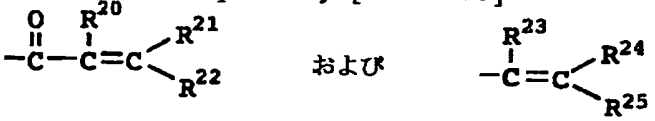


a shell -- it is one chosen from a group, R8-R19 are the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively, m is 1-5, n is 1-20, and l is 1-20] : Polysiloxane compound [which is come out of and shown], or chemical formula 2 [Formula 4]

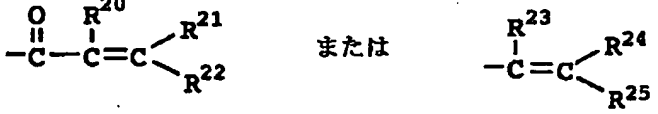


化学式2

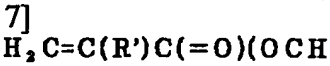
It is the alkyl in which R2' and R3' are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound among [formula, and R5 and R7 may have independently branching of carbon numbers 1-5, respectively. [Formula 5]



a shell -- it chooses from a group -- having -- at least one [however,] of R5 and the R7 [Formula 6]



It comes out, and it is, R6 is the alkyl which may have branching of a hydrogen atom or carbon numbers 1-5, and R20-R25 are [m' is 1-5, n' is 1-20, l' is 1-20, x is 1-15, and] the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively.] : The polysiloxane-polyoxyalkylene compound come out of and shown, and chemical formula 3 [Formula 7]



化学式3

[-- R' is a hydrogen atom or CH3 among a formula, R" is a hydrogen atom, -C(=O) CH=CH2, or -C (=O) C(CH3) =CH2, and z is 1-20] The macromolecule gel electrolyte which is made to harden the constituent which comes out and contains the polyethylene-glycol derivative shown and the organic solvent containing lithium salt, and is characterized by the bird clapper.

[Claim 2] The aforementioned polyethylene-glycol derivative is a macromolecule gel electrolyte according to claim 1 characterized by being or more [it is chosen from the group which consists of polyethylene glycol dimethacrylate, polyethylene-glycol diacrylate, polyethylene glycol monomethacrylate, and polyethylene-glycol monoacrylate] one.

[Claim 3] The macromolecule gel electrolyte according to claim 1 or 2 with which the content of the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or the aforementioned chemical formula 2 shown with the aforementioned chemical formula 1 is characterized by being 0.1 - 10 mass section, and for the content of the polyethylene-glycol derivative shown with the aforementioned chemical formula 3 being 0.4 - 50 mass section, and the content of the organic solvent containing lithium salt being 50 - 97 mass section on the basis of the constituent 100 mass section.

[Claim 4] The aforementioned constituent is a macromolecule gel electrolyte given in any 1 term of claims 1-3 characterized by including ethoxylation trimethylolpropane triacrylate further.

[Claim 5] The macromolecule gel electrolyte according to claim 4 with which the content of the aforementioned ethoxylation trimethylolpropane triacrylate is characterized by being 0 - 5 mass section on the basis of the constituent 100 mass section.

[Claim 6] The aforementioned constituent is a macromolecule gel electrolyte given in any 1 term of claims 1-5 characterized by including one or more polymerization initiators chosen from a benzophenone, a benzoyl peroxide, an acetyl peroxide, a lauroyl peroxide, and the group that consists of an azobisisobutyronitril in 0.1 - 5 mass section pan on the basis of the constituent 100 mass section.

[Claim 7] The aforementioned hardening is a macromolecule gel electrolyte given in any 1 term of claims 1-6 which is made by thermal polymerization, the polymerization by the electron beam, or the polymerization by UV, and is characterized by the bird clapper.

[Claim 8] The macromolecule gel electrolyte according to claim 7 characterized by polymerization temperature being 60-100 degrees C in the aforementioned thermal polymerization.

[Claim 9] It is a macromolecule gel electrolyte given in any 1 term of claims 1-8 characterized by for the aforementioned lithium salt being or more [it is chosen from the group which consists of LiClO4, LiBF4, LiPF6, LiAsF6, LiCF3SO3, and LiN(CF3SO2) 2] one, and the aforementioned organic solvent being or more [it is chosen from the group which consists of propylene carbonate, ethylene carbonate, dimethyl carbonate, methylethyl carbonate, diethyl carbonate, vinylene carbonate, triglyme, tetraglyme, and gamma-butyrolactone] one.

[Claim 10] The lithium cell possessing the case having the electrode assembly containing the

separator which intervenes between a cathode, an anode, and the aforementioned cathode and the aforementioned anode, a macromolecule gel electrolyte given in any 1 term of claims 1-9, and the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte.
[Claim 11] It is the lithium cell according to claim 10 which the aforementioned electrode assembly is formed of rolling up, and is characterized by the aforementioned case being a pouch type.
[Claim 12] The aforementioned separator is a lithium cell according to claim 10 or 11 characterized by being the sheet which combined a polyethylene sheet, a polypropylene sheet, or these.

[Translation done.]

* NOTICES *

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3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[The technical field to which invention belongs] this invention relates to the lithium cell which raised safety and reliability using the macromolecule gel electrolyte which can maintain a gel more good in a detail about the lithium cell which comes to use a macromolecule gel electrolyte and it.
[0002]
[Description of the Prior Art] Compared with a conventional nickel-cadmium battery and a conventional nickel hydrogen atom cell, the energy density and voltage per unit volume are high, the weight of a cell is also those half extent, and a lithium cell is characterized by the charge-and-discharge cycle-life property being excellent, and not having a bad influence on environment. Therefore, the lithium cell attracts the interest as a next-generation highly efficient battery, and the contribution to the formation of small lightweight and prolonged use of portable electronic equipment is expected.
[0003] A lithium cell can be divided roughly into the lithium ion battery which uses a liquid electrolyte, and the lithium ion polymer battery which uses a polymer-like electrolyte. A lithium ion battery seals an electrode assembly in the case of a cylindrical shape or a square shape. However, recently, the method of sealing an electrode assembly by the pouch attracts attention instead of using such a case. There is an advantage which thin-shape-izing and lightweight-izing of a cell are possible, and can also cut down the cost of materials by use of a pouch since a unit mass and the energy density per volume can be raised further.
[0004] Drawing 1 is the decomposition perspective diagram having shown typically an example of the lithium ion battery which used the general pouch. The lithium ion battery shown by drawing 1 consists of the electrode assembly 10 containing a cathode 11, an anode 12, and separator 13, and the case 14 which wraps and seals this electrode assembly 10. In this case, the electrode assembly 10 is formed by rolling round what inserted separator 13 between the cathode 11 and the anode 12. Furthermore, the cathode tap 15 and anode tap 15' which play the role of the electric path of the electrode assembly 10 and the exterior are pulled out from a cathode 11 and an anode 12, and form an electrode terminal 16 and 16'.
[0005] Drawing 2 is the decomposition perspective diagram having shown an example of the conventional lithium ion polymer battery typically. The lithium ion polymer battery shown by drawing 2 consists of the electrode assembly 21 containing a cathode, an anode, and separator, and the case 22 which wraps and seals the electrode assembly 21. in this case -- an electrode -- an assembly -- 21 -- having been generated -- current -- the exterior -- guiding -- a sake -- electric -- a path -- a role -- achieving -- an electrode terminal (or lead wire) -- 24 -- 24 -- ' -- a cathode -- and -- an anode -- having -- having had -- a cathode -- a tap -- 23 -- and -- an anode -- a tap -- 23 -- ' -- connecting -- having -- **** -- these -- a case -- 22 -- outside -- predetermined -- length -- extending
[0006] After putting in the electrode assemblies 10 or 21 in a case 14 or 22 and pouring in the electrolytic solution here, exposing a part of electrode-terminal 16, 16' or 24, and 24' outside, such a

lithium ion battery of drawing 1 and a lithium ion polymer battery of drawing 2 apply heat and a pressure, and are manufactured by making it paste up with the heat adhesive property matter, and sealing the marginal part of an up case, and the marginal part of a lower case.

[0007] When the boiling point uses the electrolyte containing a low organic solvent here, the phenomenon in which an electrode assembly and a pouch expand occurs and there is a problem that the reliability and the safety of a cell fall.

[0008] In order to solve such a problem, the method of stiffening a flat-surface form cell with ultraviolet rays (UV) and an electron beam, and making it, or not pouring in the electrolytic solution separately, i.e., the method of coating an electrode pipe with gel beforehand etc., is proposed (U.S. Pat. No. 5,972,539, U.S. Pat. No. 5,279,910, U.S. Pat. No. 5,437,942, and U.S. Pat. No. 5,340,368). However, by these methods, it was not able to prevent and ease with the level with which should be satisfied of an expansion phenomenon.

[0009]

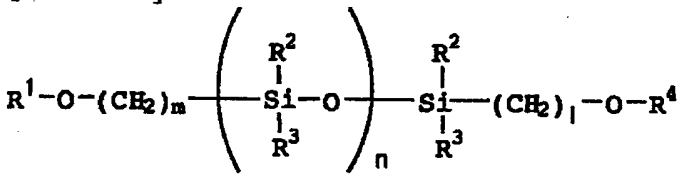
[Problem(s) to be Solved by the Invention] The purpose of this invention is offering the macromolecule gel electrolyte which can suppress the expansion phenomenon by the electrolytic solution effectively. Moreover, other purposes of this invention are offering the high lithium cell of reliability and safety using such a macromolecule gel electrolyte.

[0010]

[Means for Solving the Problem] The polyethylene oxide conventionally used in order that this invention persons may prepare a gel electrolyte, Instead of the polymer which makes a polypropylene oxide a principal chain and has functional groups, such as an acrylic, a vinyl, and an epoxy group While excelling in a physical characteristic and an electrochemical property by hardening the constituent containing the polysiloxane compound or polysiloxane-polyoxyalkylene compound containing a siloxane unit or an oxy-alkylene unit It finds out that the electrolyte which can suppress expansion effectively is obtained, and came to complete this invention.

[0011] Therefore, this invention is chemical formula 1: [0012].

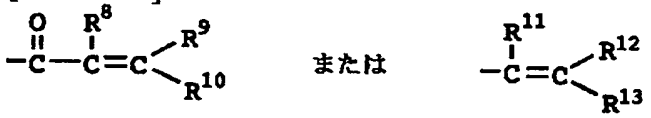
[Formula 8]



化学式1

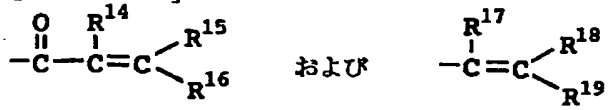
[0013] The inside of [formula and R1 are [0014].

[Formula 9]



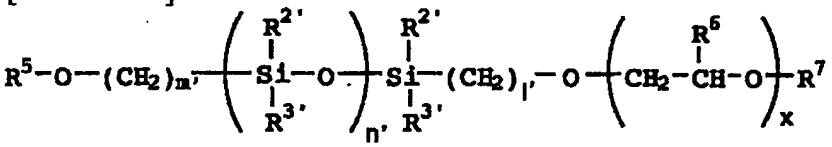
[0015] It is the alkyl in which it appears, and exists, R2 and R3 are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound, and R4 may have branching of an allyl compound and carbon numbers 1-5, and [0016].

[Formula 10]



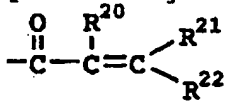
[0017] a shell -- it is one chosen from a group, R8-R19 are the alkyls which may have independently

branching of a hydrogen atom or carbon numbers 1-5, respectively, m is 1-5, n is 1-20, and l is 1-20] Polysiloxane compound [which is come out of and shown], or chemical formula 2: [0018]
[Formula 11]

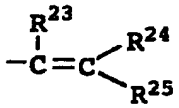


化学式2

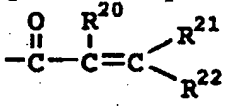
[0019] It is the alkyl in which R2' and R3' are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound among [formula, and R5 and R7 may have independently branching of carbon numbers 1-5, respectively, and [0020].
[Formula 12]



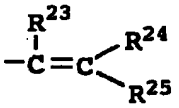
および



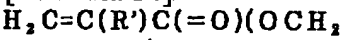
[0021] a shell -- it chooses from a group -- having -- at least one [however,] of R5 and the R7 --
[0022]
[Formula 13]



または



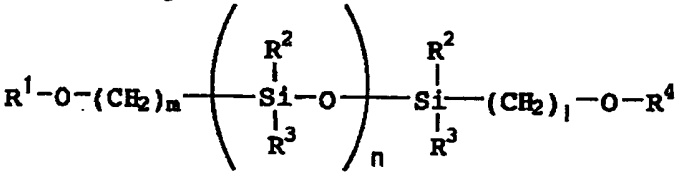
[0023] It comes out, and it is, R6 is the alkyl which may have branching of a hydrogen atom or carbon numbers 1-5, and R20-R25 are [m' is 1-5, n' is 1-20, l' is 1-20, x is 1-15, and] the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively.] The polysiloxane-polyoxyalkylene compound come out of and shown, and chemical formula 3:
[0024]
[Formula 14]



化学式3

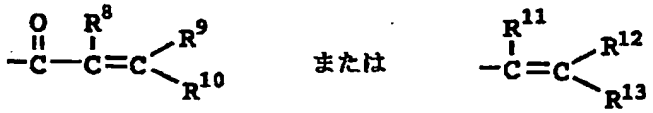
[0025] [-- R' is a hydrogen atom or CH3 among a formula, R" is a hydrogen atom, -C(=O) CH=CH2, or -C(=O) C(CH3) =CH2, and z is 1-20] It is the macromolecule gel electrolyte which is made to harden the constituent which comes out and contains the polyethylene-glycol derivative shown and the organic solvent containing lithium salt, and is characterized by the bird clapper.
[0026] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by the aforementioned polyethylene-glycol derivative being or more [it is chosen from the group which consists of polyethylene glycol dimethacrylate, polyethylene-glycol diacrylate, polyethylene glycol monomethacrylate, and polyethylene-glycol monoacrylate] one.
[0027] The content of the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or the aforementioned chemical formula 2 in which this invention is furthermore shown with the aforementioned chemical formula 1 is the aforementioned macromolecule gel electrolyte which the content of the polyethylene-glycol derivative which is 0.1 - 10 mass section and is shown with the aforementioned chemical formula 3 is 0.4 - 50 mass section, and is characterized by the content of the organic solvent containing lithium salt being 50 - 97 mass section on the basis of the constituent 100 mass section.
[0028] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized

by the aforementioned constituent containing ethoxylation trimethylolpropane triacrylate further.
[0029] Furthermore, it is the aforementioned macromolecule gel electrolyte with which, as for this invention, the content of the aforementioned ethoxylation trimethylolpropane triacrylate is characterized by being 0 - 5 mass section on the basis of the constituent 100 mass section.
[0030] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by the aforementioned constituent containing in 0.1 - 5 mass section pan one or more polymerization initiators chosen from a benzophenone, a benzoyl peroxide, an acetyl peroxide, a lauroyl peroxide, and the group that consists of an azobisisobutyronitril on the basis of the constituent 100 mass section.
[0031] Furthermore, this invention is the aforementioned macromolecule gel electrolyte which the aforementioned hardening is made by thermal polymerization, the polymerization by the electron beam, or the polymerization by UV, and is characterized by the bird clapper.
[0032] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by polymerization temperature being 60-100 degrees C in the aforementioned thermal polymerization.
[0033] Furthermore, this invention is, or more [the aforementioned lithium salt is chosen from the group which consists of LiClO4, LiBF4, LiPF6, LiAsF6, LiCF3SO3, and LiN(CF3SO2) 2] one, and the aforementioned organic solvent is the aforementioned macromolecule gel electrolyte characterized by being, or more [it is chosen from the group which consists of propylene carbonate, ethylene carbonate, dimethyl carbonate, methylethyl carbonate, diethyl carbonate, vinylene carbonate, triglyme tetraglyme, and gamma-butyrolactone] one.
[0034] Furthermore, this invention is a lithium cell possessing the case having the electrode assembly containing the separator which intervenes between a cathode, an anode, and the aforementioned cathode and the aforementioned anode, the aforementioned macromolecule gel electrolyte, and the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte.
[0035] It is the aforementioned lithium cell which the aforementioned electrode assembly is formed for this invention of rolling up, and is furthermore characterized by the aforementioned case being a pouch type.
[0036] Furthermore, this invention is the aforementioned lithium cell characterized by the aforementioned separator being the sheet which combined a polyethylene sheet, a polypropylene sheet, or these.
[0037]
[Embodiments of the Invention] The macromolecule gel electrolyte of this invention makes it come to harden the constituent containing a polysiloxane compound, a polysiloxane-polyoxyalkylene compound, a polyethylene-glycol derivative, and the organic solvent containing lithium salt. the constituent concerned -- the following and "the constituent for forming a macromolecule gel electrolyte" -- or it is only called a "constituent".
[0038] Each component first contained in a constituent is explained.
[0039] A polysiloxane compound is chemical formula 1: [0040].
[Formula 15]

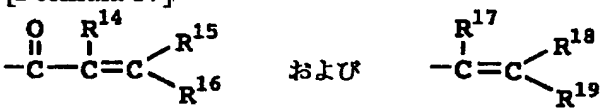


化学式 1

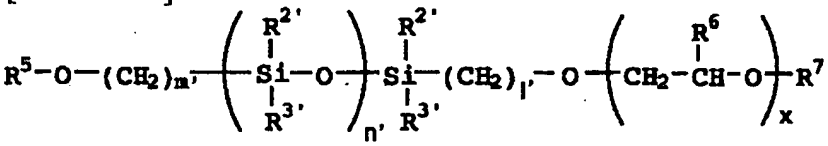
[0041] It is come out and shown. The inside of a formula and R1 are [0042].
[Formula 16]



[0043] It comes out, and it is and R8-R13 are the alkyls which may have branching of a hydrogen atom or carbon numbers 1-5. As R1, specifically, -C(=O) CH=CH2, -C(=O) C(CH3) =CH2, and -C (=O) CH=CHCH3 are mentioned, and it is -C(=O) C(CH3) =CH2 preferably.
[0044] R2 and R3 are chosen independently from the group which consists of the alkyl and phenyl which may have branching of carbon numbers 1-5, a benzyl, and an allyl compound, respectively. As R2 and R3, specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, and an isopentyl are mentioned, and they are a methyl, ethyl, a phenyl, or a benzyl preferably.
[0045] R4 is an allyl compound (-CH2 CH=CH2), the alkyl which may have branching of carbon numbers 1-5, and [0046].
[Formula 17]



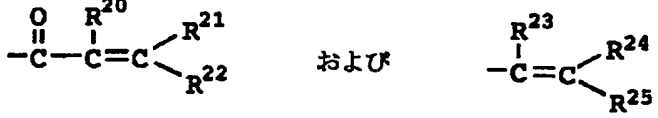
[0047] a shell -- it is one chosen from a group and R14-R19 are the alkyls which may have branching of a hydrogen atom or carbon numbers 1-5 As R4, specifically, -C(=O) C(CH3) =CH2, -C(=O) CH=CHCH3, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, and an isopentyl are mentioned, and they are a methyl and -C(=O) C(CH3) =CH2 preferably.
[0048] m is 1-5. n -- 1-20 -- it is 1-10 preferably l -- 1-20 -- it is 1-10 preferably When m, n, and l are this range, since the compound shown with a chemical formula 1 is excellent in solubility, reactivity, and the cell performance side, it is desirable.
[0049] As such a polysiloxane compound, -C(=O) C(CH3) =CH2 and R2 A methyl, [R1] The polysiloxane compound whose R4 R3 is a methyl and is a methyl, and m, n and l3, Ethyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 3 or whose 51 2 or 3n is [R4] 3 or 5 for a methyl and m, A phenyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l a methyl and m are 3 or 5 for R4, A benzyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l a methyl and m are 3 or 5 for R4, A phenyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] R4 -- ethyl and m= -- 2 or 3, and n= -- 3 or 5, and l= -- the polysiloxane compound which is 3 or 5 -- A benzyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l ethyl and m are 3 or 5 for R4, Since it excels in ********, reactivity, and the cell performance side, it is used preferably, and the polysiloxane compound a methyl and whose R4 are [R1 / -C(=O) C(CH3) =CH2 and R2] a methyl, and m, n and l3 also especially in these for a methyl and R3 is desirable.
[0050] Next, a polysiloxane-polyoxyalkylene compound is explained. The compound concerned is chemical formula 2: [0051].
[Formula 18]



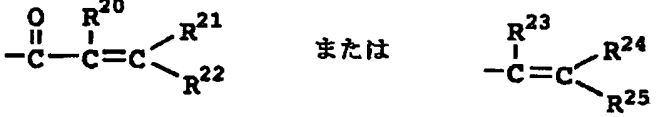
化学式2

[0052] It is come out and shown. R2' and R3' are chosen from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound among a formula. Specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl,

sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned, and they are a methyl, ethyl, a phenyl, or a benzyl preferably.
[0053] R5 and R7 are the alkyl which may have branching of carbon numbers 1-5, and [0054] independently, respectively.
[Formula 19]



[0055] a shell -- it chooses from a group -- having -- at least one [however,] of R5 and the R7 -- [0056]
[Formula 20]



[0057] It comes out, and it is and R20-R25 are the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively. As R5 or R7, specifically, -C(=O) C(CH3) =CH2, -C(=O) CH=CHCH3, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned, and they are -C(=O) C(CH3) =CH2 or a methyl preferably.
[0058] R6 is the alkyl which may have branching of a hydrogen atom or carbon numbers 1-5, and, specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned as this alkyl. It is a hydrogen atom preferably as R6.
[0059] m' is 1-5. n' is 1-20 and is 1-10 preferably. l' is 1-20 and is 1-10 preferably. x is 1-15. m -- ' -- n -- ' -- l -- ' -- and -- x -- this -- the range -- it is -- the time -- a chemical formula -- two -- being shown -- having -- a compound -- solubility -- reactivity -- and -- a cell -- a performance -- a field -- excelling -- **** -- a sake -- being desirable .
[0060] As a polysiloxane-polyoxyalkylene compound shown with such a chemical formula 2 -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a methyl and R3'] [a methyl and R5] R7 A methyl, m', the polysiloxane-polyoxyalkylene compound whose x n' and l' are 3 and is 5, -C(=O) C (CH3) =CH2 and R6 A hydrogen atom, [R2'] [ethyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a benzyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a phenyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C (CH3) =CH2 and R6 A hydrogen atom, [R2'] [a benzyl and R3'] [a methyl and R5] Since the polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m' is excellent in solubility, reactivity, and the cell performance side, it is used preferably. these -- inside -- **** -- especially -- R -- two -- ' -- a methyl -- R -- three -- ' -- a methyl -- R -- five - C (=O) -- C (CH3) -- = -- CH -- two -- R -- six -- a hydrogen atom -- R -- seven -- a methyl -- m -- ' -- n -- ' -- and -- l -- ' -- three -- x -- five -- it is -- a polysiloxane -- a compound -- being desirable .
[0061] As for the content of the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with a chemical formula 1, it is desirable that it is 0.1 - 10 mass section on the basis of the constituent 100 mass section. The effect of a performance improvement of the cell according to addition of these compounds when a content is under the 0.1 mass section here is low, and when exceeding 10 mass sections, there is a possibility that the performance of a cell may fall.

[0062] Although the polysiloxane compound shown with a chemical formula 1 can make for example, a dihydroxy end polysiloxane and an acryloyl chloride able to react and can manufacture a polysiloxane compound, it is not limited to this but can be manufactured by the method of this business world common knowledge. Similarly, the polysiloxane-polyoxyalkylene compound shown with a chemical formula 2 can be manufactured by various methods. For example, although an acryloyl chloride can be made to be able to react to the polysiloxane-polyoxyalkylene which a dihydroxy end polysiloxane and ethylene oxide are made to react, and is manufactured and a polysiloxane-polyoxyalkylene compound can be manufactured, it is not limited to this but can manufacture by the method of this business world common knowledge.

[0063] Next, a polyethylene-glycol derivative is explained. The polyethylene-glycol derivative concerned is chemical formula 3: [0064].

[Formula 21]

$$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$$

化学式3

[0065] It is come out and shown. R' is a hydrogen atom or CH3 among a formula, R" is a hydrogen atom, -C(=O) CH=CH2, or -C(=O) C(CH3) =CH2, and z is 1-20. Polyethylene glycol dimethacrylate, polyethylene-glycol diacrylate, polyethylene glycol monomethacrylate, polyethylene-glycol monoacrylate, etc. are mentioned preferably as such a polyethylene-glycol derivative. These may be used independently, respectively, and it may mix and they may be used. By using especially polyethylene glycol dimethacrylate, polyethylene glycol monomethacrylate, or those mixture, since the performance of a cell can be raised, it is desirable.

[0066] The range of the content of the polyethylene-glycol derivative concerned has desirable 0.4 - 50 mass section on the basis of the constituent 100 mass section. When a content is under the 0.4 mass section here, hardening reactivity falls, and when exceeding 50 mass sections, there is a possibility that the performance of a cell may fall. However, since the range of the content of a polyethylene-glycol derivative is various by what is used, it is not this limitation. Hereafter, the range of a desirable content is described concretely. For example, as for the content of polyethylene glycol dimethacrylate, it is desirable that it is 0.5 - 50 mass section on the basis of the constituent 100 mass section, and, as for the content of polyethylene glycol monomethacrylate, it is desirable that it is 0.4 - 50 mass section on the basis of the constituent 100 mass section.

[0067] Next, the organic solvent containing the lithium salt of this invention is explained. It is desirable that it is or more [it is chosen from the group which consists of a lithium perchlorate (LiClO4), a 4 boron-fluoride lithium (LiBF4), a 6 fluoride / phosphorus / lithium (LiPF6), a 6 fluoride arsenic lithium (LiAsF6), a 3 fluoride methansulfonic acid lithium (LiCF3SO3), and a RICHIUMUBISU trifluoromethane sulfonyl amide (LiN2 (CF3SO2)) as lithium salt] one. It is LiPF6 especially preferably. Of course, well-known lithium salt is usable at the technical field concerned besides the above.

[0068] It is desirable that it is or more [it is chosen from the group which consists of propylene carbonate, ethylene carbonate, dimethyl carbonate, methylethyl carbonate, diethyl carbonate, BINIRUREN carbonate, triglyme, tetraglyme, and gamma-butyrolactone as an organic solvent] one. Especially, preferably, it is ethylene carbonate (EC), dimethyl carbonate (DMC), and dimethyl carbonate (DEC), and these may be used independently, and you may mix and use by the suitable volume ratio. Of course, a well-known organic solvent is usable at the technical field concerned besides the above.

[0069] As for the content of the organic solvent containing such lithium salt, it is desirable that it is 50 - 97 mass section on the basis of the constituent 100 mass section. And as for the content of lithium salt, it is desirable that it is 0.5-3 mols to organic-solvent 1L. When the content of an organic solvent and lithium salt separates from the aforementioned range here, there is a possibility that the performance of a cell may fall.

[0070] As for the constituent of this invention, it is still more desirable that ethoxylation trimethylolpropane triacrylate (ethoxylated trimethylol propane triacrylate) is included. The hardening reaction of a constituent can be promoted by including such a compound. The content of ethoxylation trimethylolpropane triacrylate has a possibility that the performance of a cell may fall, when it is desirable that it is 0 - 5 mass section and it exceeds 5 mass sections on the basis of the constituent 100 mass section here.

[0071] The constituent for forming the macromolecule gel electrolyte of this invention can be obtained by mixing the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with the chemical formula 1 described above, a polyethylene-glycol derivative, and the organic solvent containing lithium salt.

[0072] Furthermore, as for the aforementioned constituent, it is desirable that one or more polymerization initiators chosen from a benzophenone, a benzoyl peroxide, an acetyl peroxide, a lauroyl peroxide, and the group that consists of an azobisisobutyronitril are included, and, generally especially an azobisisobutyronitril is used. Generally, on the basis of the constituent 100 mass section, although the content of a polymerization initiator is 0.1 - 5 mass section, it should choose each suitable content according to the kind here.

[0073] The macromolecule gel electrolyte of this invention is manufactured by hardening the constituent containing an above-mentioned component. Hardening is desirable and thermal polymerization, the polymerization by the electron beam, or the polymerization by UV is used. Each polymerization method is explained in detail below.

[0074] Thermal polymerization is performed by processing a constituent in predetermined polymerization time in the oven adjusted by polymerization temperature. In the case of less than 60 degrees C, as for polymerization temperature, it is desirable that it is 60-100 degrees C, and there is a possibility that a polymerization may become inadequate, and if 100 degrees C is exceeded on the other hand, an expansion phenomenon may produce it here. carrying out the polymerization of the polymerization time for example, within a case -- or although it is not limited since it is required to adjust suitably by whether a polymerization is carried out in the state where it cast for the film, generally it is about 4 hours

[0075] The polymerization by the electron beam is made when predetermined carries out time irradiation of the electron beam at a constituent. The output of this electron beam is usually 1.5eV or more, and in the case of less than 1.5eV, the long setting time is needed or it has fear, like hardening becomes inadequate here. Generally, as for time to irradiate an electron beam, it is desirable that it is 30 seconds or more, and when time is less than 30 seconds, it has a possibility that a polymerization may become inadequate. The polymerization by such electron beam can shorten reaction time compared with above-mentioned thermal polymerization, and has the advantage of not needing a polymerization initiator further.

[0076] The polymerization by UV is made when predetermined carries out time irradiation of the UV at a constituent. Generally, the wavelength of this UV has a possibility that a polymerization may become inadequate, when it is desirable that it is 250-365nm and it separates from this range here. Generally, as for time to irradiate UV, it is desirable that it is 30 seconds or more, and when time is less than 30 seconds, it has a possibility that a polymerization may become inadequate. The polymerization by such UV has the advantage that reaction time can be shortened compared with above-mentioned thermal polymerization.

[0077] Next, how to manufacture a macromolecule gel electrolyte using the constituent mentioned above is explained. It prepares within the limits of the content which mentioned each component above first. These contents should be chosen the optimal in consideration of the performance of the electrochemical stability of the lithium cell manufactured, and a cell etc. Next, the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with a chemical formula 1, the polyethylene-glycol derivative shown with a chemical formula 3, and ethoxylation trimethylolpropane triacrylate are mixed. Into this mixture, the above-mentioned

polymerization initiator is added further if needed, the organic solvent which contains lithium salt further is added and stirred into it, and a uniform constituent is obtained. The obtained constituent is hardened by the thermal polymerization mentioned above, the polymerization by the electron beam, or the polymerization by UV.

[0078] Next, the lithium cell using the macromolecule gel electrolyte of this invention is explained. The lithium cell of this invention possesses the case having the electrode assembly containing the separator which intervenes between a cathode, an anode, and the aforementioned cathode and the aforementioned anode, the macromolecule gel electrolyte of this invention mentioned above, and the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte.

[0079] Explanation of the cathode active material constituent and anode active material constituent which are first used as a cathode or an anode uses preferably for a cathode the cathode active material constituent which generally consists of lithium content oxides, such as LiCoO2. The anode active material constituent which generally contains carbon, graphite, etc. is preferably used for an anode, and, generally a mezzo-soprano carbon fiber is desirable especially as carbon.

[0080] Furthermore, these active material constituent may contain the electric conduction agent, the binder, and the solvent suitably. As an electric conduction agent, although not limited to these, carbon black etc. is used preferably. Since the conductivity of an electrode active material layer falls when it is desirable that it is 1 - 20 mass section and a content exceeds 20 mass sections here on the basis of the electrode active material (for example, LiCoO2) 100 mass section, and the content of an electrode active material becomes low relatively in being under 1 mass section, the content of an electric conduction agent is not desirable here.

[0081] As a binder, a vinylidene fluoride-hexafluoropropylene copolymer (VdF/HFP copolymer), poly vinylidene fluoride, a polyacrylonitrile, the poly methyl methacrylic rate, or its mixture is used preferably, and, generally especially poly vinylidene fluoride is desirable. As for the content of a binder, it is desirable that it is 2 - 30 mass section on the basis of the electrode active material 100 mass section. When a content is the aforementioned range here, it excels in the bonding strength of the electrode active material to an electrode charge collector.

[0082] As a solvent, if generally used for a lithium cell, all are usable, for example, an acetone and N-methyl pyrrolidone are mentioned, and it combines with other components, and is chosen suitably.

[0083] Although separator will not be restricted especially if used in the field concerned, it is desirable that it is the sheet which combined a polyethylene sheet, a polypropylene sheet, or these. It is preferably used by the reason especially for being easy to roll round polyethylene separator, and polypropylene / polyethylene / three layer separator of polypropylene. Specifically, the polyethylene separator by the morning-sun chemical-industry company is used.

[0084] Although all are usable if it is the type used in this industry, since especially a pouch type can prevent external disclosure of a gel electrolyte good, the case having the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte containing such a cathode, an anode, and separator is desirable.

[0085] Next, the manufacture method of the lithium cell of this invention is explained. First, an electrode active material layer is formed on a charge collector using an above-mentioned anode active material constituent or an above-mentioned cathode active material constituent, and a cathode electrode board and an anode electrode board are produced. An electrode active material layer is formed here by the method of coating each active material constituent directly on a charge collector, and the method of carrying out the lamination of the film of the active material constituent obtained from this base material by exfoliating after coating and drying each active material constituent in the base material upper part separately with a doctor blade on a charge collector. If an active material layer can be supported, such a base material can use all, for example, polyethylene terephthal rate films, such as a Mylar film (Du Pont make), are suitable for it.

[0086] Next, separator is inserted between the cathode electrode boards and anode electrode boards which were obtained, and an electrode assembly is formed in it. Even if it forms the assembly

concerned by rolling up using the jelly roll method (drawing 1), you may form it in the Bayh cellular structure here (drawing 2). Thus, the obtained assembly is installed in a case.
[0087] Next, the macromolecule gel electrolyte of this invention is formed by preparing the constituent for forming the macromolecule gel electrolyte of this invention in a case, and hardening it. The method of hardening the case which poured in the constituent for forming the macromolecule gel electrolyte of this invention into the case as one method, sealed, and was acquired by the polymerization by the electron beam mentioned above or the polymerization by UV is mentioned. Thus, when carrying out the polymerization of the constituent within a case, the effect by thermal polymerization can stiffen a constituent most efficiently.
[0088] After casting the constituent for forming the electrolyte of this invention in either an anode electrode board or a cathode electrode board and the front face of both these with a doctor blade as other methods, the method of hardening by the polymerization by the electron beam mentioned above or the polymerization by UV is also desirable. Thus, the lithium cell of this invention can be obtained.
[0089] The lithium cell of this invention is applicable to all of lithium rechargeable batteries, such as a lithium primary cell, a lithium ion polymer battery, or a lithium ion battery.
[0090]
[Example] Hereafter, an example is given and this invention is explained more to a detail.
[0091] Added poly vinylidene fluoride 15g to <production of macromolecule gel electrolyte and lithium cell using it> example 1 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and acetylene black (tradename : super - P, product made from MMM) 15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was produced.
[0092] The aforementioned cathode active material constituent was coated using the doctor blade which has the gap of 320 micrometers on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, it dried and the unit cathode electrode board was produced.
[0093] On the other hand, poly vinylidene fluoride 50g was added to acetone 600ml, mix for 2 hours, it was made to dissolve with a ball mill, mixture was obtained, mezzo-soprano carbon fiber 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was produced.
[0094] The aforementioned anode active material constituent was coated using the doctor blade which has the gap of 420 micrometers on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, it dried and the unit anode electrode board was produced.
[0095] Polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm] and a thickness of 18 micrometers was made to intervene between the aforementioned cathode electrode board and the aforementioned anode electrode board, this was rolled round by the jelly roll method, and the electrode assembly was produced. This electrode assembly was put into the pouch.
[0096] On the other hand, it sets in a chemical formula 1. a methyl and R1 -C(=O) C(CH3) =CH 2 and m, [R4 R2, and R3] 0.2g [of polysiloxane compounds], and polyethylene-glycol JIMETA krill rate 1.8g whose l is n and3, Polyethylene-glycol monochrome methacrylic rate 0.5g, ethoxylation trimethylolpropane triacrylate 0.5g, Azobisisobutyronitril 0.1g, and (ethylene carbonate EC) / dimethyl carbonate (DMC) / dimethyl carbonate (DEC) = 30g of organic solvents which are 3:3:1 (volume ratio) and contain LiPF6 1M is mixed. The constituent for forming a macromolecule gel electrolyte was prepared. 3g of this constituent was poured into the pouch containing the electrode assembly obtained by the above-mentioned method, and it was sealed. The lithium cell of this invention was completed by as a result processing an object in the oven adjusted at 80 degrees C for 4 hours.
[0097] Added poly vinylidene fluoride 15g to example 2 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. After adding LiCoO2470g and super-P

(product made from MMM)15g into this mixture, this was mixed for 5 hours and the cathode active material constituent was produced.

[0098] The doctor blade which has the gap of 320 micrometers for the obtained cathode active material constituent was used, and it coated on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, and it dried and the unit cathode electrode board was produced.

[0099] Then, 0.2g [of the same polysiloxane compounds as an example 1] and polyethylene-glycol JIMETA krill rate 1.8g, polyethylene-glycol monochrome methacrylic rate 0.5g, ethoxylation trimethylolpropane triacrylate 0.5g, benzophenone 0.1g, and 30g of organic solvents which are EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M were mixed, and the constituent for forming a macromolecule gel electrolyte was manufactured. After using and casting a doctor blade for the cathode electrode board which manufactured this constituent above, 365nm UV is irradiated for 1.5 hours, and was stiffened, and the cathode electrode in which the macromolecule gel electrolyte was formed was obtained.

[0100] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and the constituent was obtained. Mezzo-soprano carbon fiber 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0101] The doctor blade which has the gap of 420 micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0102] After making polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm], and a thickness of 18 micrometers intervene between the aforementioned cathode electrode boards and the aforementioned anode electrode boards with which the macromolecule gel electrolyte was formed, this was rolled round by the jelly roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch and the lithium cell was completed.

[0103] If it removed having made the anode electrode board cast and harden the constituent for forming an example 3 macromolecule gel electrolyte instead of a cathode electrode board, the lithium cell was produced by the same method as an example 2.

[0104] If it removed having cast the constituent for forming an example 4 macromolecule gel electrolyte for both the cathode electrode board and the anode electrode board, the lithium cell was produced by the same method as an example 2.

[0105] Added poly vinylidene fluoride 15g to example 5 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and super-P (product made from MMM)15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was prepared.

[0106] The doctor blade which has the gap of 320 micrometers for the aforementioned cathode active material constituent was used, and it coated on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, and it dried and the unit cathode electrode board was produced.

[0107] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. Mezzo-soprano carbon fiber (MCF) 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0108] The doctor blade which has the gap of 420 micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0109] After making polyethylene separator (morning-sun chemical-industry company) with 18 micrometers [in thickness], and a width of face of 5.25cm intervene between the aforementioned cathode electrode board and the aforementioned anode electrode board, this was rolled round by the ZERU roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch.

[0110] On the other hand, it sets in a chemical formula 2. -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2', R3', and R7] [a methyl and R5] 3 and x 0.2g of 5 polysiloxane-polyoxyalkylene compounds, [m', n', and l'] Polyethylene-glycol JIMETAKURIRURE - TO1.8g, polyethylene-glycol monochrome methacrylic rate 1g, Ethoxylation trimethylolpropane triacrylate 0.05g, azobisisobutyronitril 0.01g, and 30g of organic solvents which are EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M are mixed. The constituent for forming a macromolecule gel electrolyte was manufactured. 3g of this constituent was poured into the pouch cell obtained in the example 1, and this was sealed. Then, the lithium cell was produced by processing join fruit in the oven adjusted at 80 degrees C for 4 hours.

[0111] Added poly vinylidene fluoride 15g to example 6 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and super-P (product made from MMM)15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was prepared.

[0112] The aforementioned cathode active material constituent was coated using the doctor blade which has the gap of 320 micrometers on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, it dried and the unit cathode electrode board was produced.

[0113] Then, 0.2g [of the same polysiloxane-polyoxyalkylene compounds as an example 5] and polyethylene-glycol JIMETA krill rate 1.8g, polyethylene-glycol monochrome methacrylic rate 1g, ethoxylation trimethylolpropane triacrylate 0.05g, benzophenone 0.01g, and 30g of organic solvents which are EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M were mixed, and the constituent for forming a macromolecule gel electrolyte was prepared. After using and casting a doctor blade for the cathode electrode board which manufactured this constituent above, UV with a wavelength of 365nm was irradiated for 1.5 hours, and was stiffened.

[0114] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. Mezzo-soprano carbon fiber (MCF) 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0115] The doctor blade which has the gap of 420 micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0116] After making polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm], and a thickness of 18 micrometers intervene between the cathode electrode boards and anode electrode boards with which the macromolecule gel electrolyte was formed, this was rolled round by the jelly roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch and the lithium cell was produced.

[0117] If it removed having cast the constituent for forming an example 7 macromolecule gel electrolyte for the anode electrode board instead of the cathode electrode board, the lithium cell was produced by the same method as an example 6.

[0118] If it removed having cast the constituent for forming an example 8 macromolecule gel electrolyte for both the cathode electrode board and the anode electrode board, the lithium cell was produced by the same method as an example 6.

[0119] Instead of the constituent for forming the macromolecule gel electrolyte of the example this invention of comparison, it is 1M. If it removed having used the mixed solution (Ube Industries, Ltd. make) which contains LiPF6 and EC/DMC/DEC by the volume ratio 3:3:4, the lithium cell was

produced by the same method as an example 1.
[0120] The lithium cell of the <characterization of cell> test-method examples 1-8 and the example of comparison evaluated reliability and safety by the life characteristic test, the penetration examination, the expansion test at the time of elevated-temperature neglect (85 degrees C), and the liquid spill examination under pressure 40 kgf/cm2.
[0121] The life characteristic test was made by observing the volume change of the cell according charge and discharge to a 100 cycle deed and it using the lithium cell of examples 1-8 and the example 1 of comparison. This evaluation made it desirable for there to be few volume changes.
[0122] The penetration experiment made the nail with a diameter of 5mm penetrate in the direction perpendicular to the major axis of after 3-hour charge and a cell in the center with the current of 0.2C, and investigated the existence of the ignition phenomenon of a cell, and a rupture phenomenon.
[0123] After leaving an expansion test at 85 degrees C after 3-hour charge in the current of 0.2C for 4 hours, it measured and evaluated the thickness of a cell. The thickness of the cell after neglect presupposed that it is good, when initial thickness became 110% or less.
[0124] The disclosure examination investigated the existence of a liquid spill, after pressurizing the cell for 10 seconds by the pressure of 40 kgf/cm2.
[0125] Test-result drawing 3 is a graph which shows the life property of the lithium cell of an example 1 and the example of comparison. According to this, the volume decrease of the cell of an example 1 was almost of the same grade as the example of comparison. Moreover, in the penetration examination, neither ignition nor rupture takes place, but is maintaining 110% of the initial thickness of a cell also in the expansion test further, and did not carry out a liquid spill from inside in the disclosure examination further. Since examples 2-8 also showed the almost same result, they were found by that the lithium cell of this invention has the outstanding life property.
[0126] As mentioned above, since the lithium cell of examples 1-8 can maintain the gel excellent in the electrolytic solution, it turns out that expansion of external disclosure of the electrolytic solution, the electrode assembly by the electrolytic solution, or a pouch can be suppressed, and it has the reliability and the safety superior to the cell of the example of comparison.
[0127]
[Effect of the Invention] By hardening the constituent containing the polysiloxane compound or polysiloxane-polyoxyalkylene compound containing a siloxane unit or an oxy-alkylene unit, by the bird clapper, the macromolecule gel electrolyte of this invention can offer the lithium cell which can suppress expansion effectively while it is excellent in a physical characteristic and an electrochemical property. Since expansion of an electrolytic external disclosure, the electrode assembly by the electrolytic solution, or a pouch is suppressed, the lithium cell obtained using the electrolyte concerned has high reliability and high safety.

[Translation done.]

* NOTICES *

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2. **** shows the word which can not be translated.
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TECHNICAL FIELD

[The technical field to which invention belongs] this invention relates to the lithium cell which raised safety and reliability using the macromolecule gel electrolyte which can maintain a gel more good in a detail about the lithium cell which comes to use a macromolecule gel electrolyte and it.

[Translation done.]

* NOTICES *

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PRIOR ART

[Description of the Prior Art] Compared with a conventional nickel-cadmium battery and a conventional nickel hydrogen atom cell, the energy density and voltage per unit volume are high, the weight of a cell is also those half extent, and a lithium cell is characterized by the charge-and-discharge cycle-life property being excellent, and not having a bad influence on environment. Therefore, the lithium cell attracts the interest as a next-generation highly efficient battery, and the contribution to the formation of small lightweight and prolonged use of portable electronic equipment is expected.

[0003] A lithium cell can be divided roughly into the lithium ion battery which uses a liquid electrolyte, and the lithium ion polymer battery which uses a polymer-like electrolyte. A lithium ion battery seals an electrode assembly in the case of a cylindrical shape or a square shape. However, recently, the method of sealing an electrode assembly by the pouch attracts attention instead of using such a case. There is an advantage which thin-shape-izing and lightweight-izing of a cell are possible, and can also cut down the cost of materials by use of a pouch since a unit mass and the energy density per volume can be raised further.

[0004] Drawing 1 is the decomposition perspective diagram having shown typically an example of the lithium ion battery which used the general pouch. The lithium ion battery shown by drawing 1 consists of the electrode assembly 10 containing a cathode 11, an anode 12, and separator 13, and the case 14 which wraps and seals this electrode assembly 10. In this case, the electrode assembly 10 is formed by rolling round what inserted separator 13 between the cathode 11 and the anode 12. Furthermore, the cathode tap 15 and anode tap 15' which play the role of the electric path of the electrode assembly 10 and the exterior are pulled out from a cathode 11 and an anode 12, and form an electrode terminal 16 and 16'.

[0005] Drawing 2 is the decomposition perspective diagram having shown an example of the conventional lithium ion polymer battery typically. The lithium ion polymer battery shown by drawing 2 consists of the electrode assembly 21 containing a cathode, an anode, and separator, and the case 22 which wraps and seals the electrode assembly 21. in this case -- an electrode -- an assembly -- 21 -- having been generated -- current -- the exterior -- guiding -- a sake -- electric -- a path -- a role -- achieving -- an electrode terminal (or lead wire) -- 24 -- 24 -- ' -- a cathode -- and -- an anode -- having -- having had -- a cathode -- a tap -- 23 -- and -- an anode -- a tap -- 23 -- ' -- connecting -- having -- **** -- these -- a case -- 22 -- outside -- predetermined -- length -- extending

[0006] After putting in the electrode assemblies 10 or 21 in a case 14 or 22 and pouring in the electrolytic solution here, exposing a part of electrode-terminal 16, 16' or 24, and 24' outside, such a lithium ion battery of drawing 1 and a lithium ion polymer battery of drawing 2 apply heat and a pressure, and are manufactured by making it paste up with the heat adhesive property matter, and sealing the marginal part of an up case, and the marginal part of a lower case.

[0007] When the electrolyte which contains an organic solvent with the low boiling point here is used, the phenomenon in which an electrode assembly and a pouch expand occurs and there is a problem that the reliability and the safety of a cell fall.

[0008] In order to solve such a problem, the method of stiffening a flat-surface form cell with ultraviolet rays (UV) and an electron beam, and making it, or not pouring in the electrolytic solution separately, i.e., the method of coating an electrode pipe with gel beforehand etc., is proposed (U.S. Pat. No. 5,972,539, U.S. Pat. No. 5,279,910, U.S. Pat. No. 5,437,942, and U.S. Pat. No. 5,340,368).

However, by these methods, it was not able to prevent and ease with the level with which should be satisfied of an expansion phenomenon.

[Translation done.]

* NOTICES *

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EFFECT OF THE INVENTION

[Effect of the Invention] By hardening the constituent containing the polysiloxane compound or polysiloxane-polyoxyalkylene compound containing a siloxane unit or an oxy-alkylene unit, by the bird clapper, the macromolecule gel electrolyte of this invention can offer the lithium cell which can suppress expansion effectively while it is excellent in a physical characteristic and an electrochemical property. Since expansion of an electrolytic external disclosure, the electrode assembly by the electrolytic solution, or a pouch is suppressed, the lithium cell obtained using the electrolyte concerned has high reliability and high safety.

[Translation done.]

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TECHNICAL PROBLEM

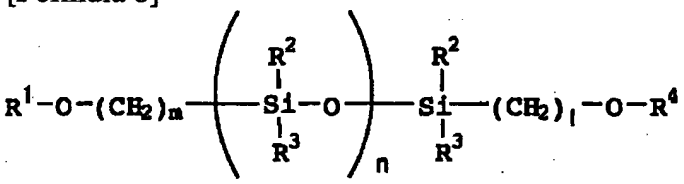
[Problem(s) to be Solved by the Invention] The purpose of this invention is offering the macromolecule gel electrolyte which can suppress the expansion phenomenon by the electrolytic solution effectively. Moreover, other purposes of this invention are offering the high lithium cell of reliability and safety using such a macromolecule gel electrolyte.

[Translation done.]

* NOTICES *

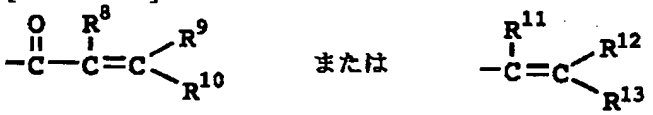
MEANS

[Means for Solving the Problem] The polyethylene oxide conventionally used in order that this invention persons may prepare a gel electrolyte, Instead of the polymer which makes a polypropylene oxide a principal chain and has functional groups, such as an acrylic, a vinyl, and an epoxy group While excelling in a physical characteristic and an electrochemical property by hardening the constituent containing the polysiloxane compound or polysiloxane-polyoxyalkylene compound containing a siloxane unit or an oxy-alkylene unit It finds out that the electrolyte which can suppress expansion effectively is obtained, and came to complete this invention.
[0011] Therefore, this invention is chemical formula 1: [0012].
[Formula 8]

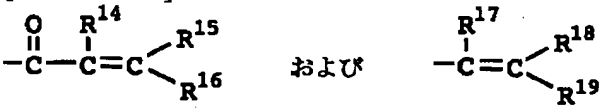


化学式1

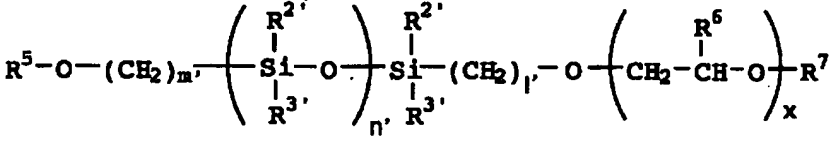
[0013] The inside of [formula and R1 are [0014].
[Formula 9]



[0015] It is the alkyl in which it appears, and exists, R2 and R3 are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound, and R4 may have branching of an allyl compound and carbon numbers 1-5, and [0016].
[Formula 10]



[0017] a shell -- it is one chosen from a group, R8-R19 are the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively, m is 1-5, n is 1-20, and l is 1-20] Polysiloxane compound [which is come out of and shown], or chemical formula 2: [0018]
[Formula 11]

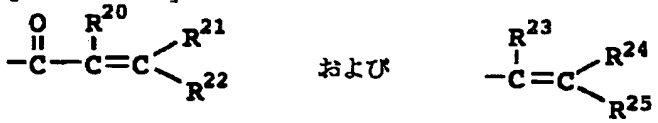


化学式2

[0019] It is the alkyl in which R2' and R3' are chosen as from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl,

and an allyl compound among [formula, and R5 and R7 may have independently branching of carbon numbers 1-5, respectively, and [0020].

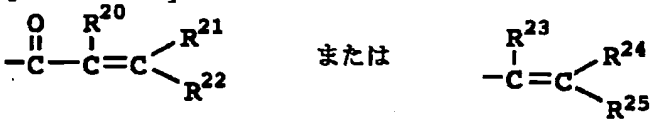
[Formula 12]



[0021] a shell -- it chooses from a group -- having -- at least one [however,] of R5 and the R7 --

[0022]

[Formula 13]



[0023] It comes out, and it is, R6 is the alkyl which may have branching of a hydrogen atom or carbon numbers 1-5, and R20-R25 are [m' is 1-5, n' is 1-20, l' is 1-20, x is 1-15, and] the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively.] The polysiloxane-polyoxyalkylene compound come out of and shown, and chemical formula 3:

[0024]

[Formula 14]

$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$

化学式3

[0025] [-- R' is a hydrogen atom or CH3 among a formula, R" is a hydrogen atom, -C(=O) CH=CH2, or -C(=O) C(CH3) =CH2, and z is 1-20] It is the macromolecule gel electrolyte which is made to harden the constituent which comes out and contains the polyethylene-glycol derivative shown and the organic solvent containing lithium salt, and is characterized by the bird clapper.

[0026] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by the aforementioned polyethylene-glycol derivative being or more [it is chosen from the group which consists of polyethylene glycol dimethacrylate, polyethylene-glycol diacrylate, polyethylene glycol monomethacrylate, and polyethylene-glycol monoacrylate] one.

[0027] The content of the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or the aforementioned chemical formula 2 in which this invention is furthermore shown with the aforementioned chemical formula 1 is the aforementioned macromolecule gel electrolyte which the content of the polyethylene-glycol derivative which is 0.1 - 10 mass section and is shown with the aforementioned chemical formula 3 is 0.4 - 50 mass section, and is characterized by the content of the organic solvent containing lithium salt being 50 - 97 mass section on the basis of the constituent 100 mass section.

[0028] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by the aforementioned constituent containing ethoxylation trimethylolpropane triacrylate further.

[0029] Furthermore, it is the aforementioned macromolecule gel electrolyte with which, as for this invention, the content of the aforementioned ethoxylation trimethylolpropane triacrylate is characterized by being 0 - 5 mass section on the basis of the constituent 100 mass section.

[0030] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by the aforementioned constituent containing in 0.1 - 5 mass section pan one or more polymerization initiators chosen from a benzophenone, a benzoyl peroxide, an acetyl peroxide, a lauroyl peroxide, and the group that consists of an azobisisobutyronitril on the basis of the constituent 100 mass section.

[0031] Furthermore, this invention is the aforementioned macromolecule gel electrolyte which the aforementioned hardening is made by thermal polymerization, the polymerization by the electron beam, or the polymerization by UV, and is characterized by the bird clapper.

[0032] Furthermore, this invention is the aforementioned macromolecule gel electrolyte characterized by polymerization temperature being 60-100 degrees C in the aforementioned thermal

polymerization.

[0033] Furthermore, this invention is, or more [the aforementioned lithium salt is chosen from the group which consists of LiClO4, LiBF4, LiPF6, LiAsF6, LiCF3SO3, and LiN(CF3SO2) 2] one, and the aforementioned organic solvent is the aforementioned macromolecule gel electrolyte characterized by being, or more [it is chosen from the group which consists of propylene carbonate, ethylene carbonate, dimethyl carbonate, methylethyl carbonate, diethyl carbonate, vinylene carbonate, triglyme tetraglyme, and gamma-butyrolactone] one.

[0034] Furthermore, this invention is a lithium cell possessing the case having the electrode assembly containing the separator which intervenes between a cathode, an anode, and the aforementioned cathode and the aforementioned anode, the aforementioned macromolecule gel electrolyte, and the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte.

[0035] It is the aforementioned lithium cell which the aforementioned electrode assembly is formed for this invention of rolling up, and is furthermore characterized by the aforementioned case being a pouch type.

[0036] Furthermore, this invention is the aforementioned lithium cell characterized by the aforementioned separator being the sheet which combined a polyethylene sheet, a polypropylene sheet, or these.

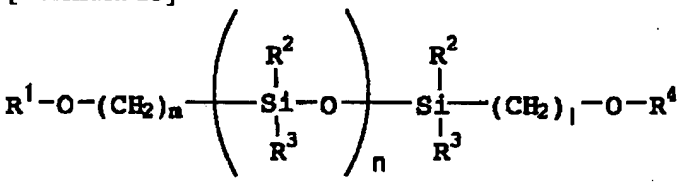
[0037]

[Embodiments of the Invention] The macromolecule gel electrolyte of this invention makes it come to harden the constituent containing a polysiloxane compound, a polysiloxane-polyoxyalkylene compound, a polyethylene-glycol derivative, and the organic solvent containing lithium salt. the constituent concerned -- the following and "the constituent for forming a macromolecule gel electrolyte" -- or it is only called a "constituent"

[0038] Each component first contained in a constituent is explained.

[0039] A polysiloxane compound is chemical formula 1: [0040].

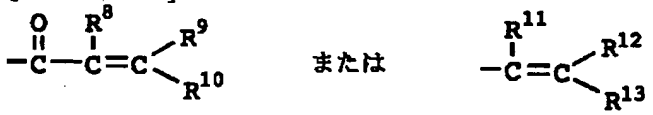
[Formula 15]



化学式1

[0041] It is come out and shown. The inside of a formula and R1 are [0042].

[Formula 16]

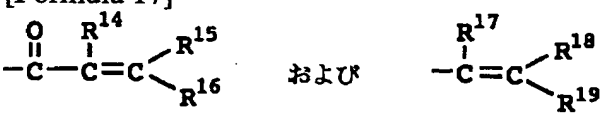


[0043] It comes out, and it is and R8-R13 are the alkyls which may have branching of a hydrogen atom or carbon numbers 1-5. As R1, specifically, -C(=O) CH=CH2, -C(=O) C(CH3) =CH2, and -C (=O) CH=CHCH3 are mentioned, and it is -C(=O) C(CH3) =CH2 preferably.

[0044] R2 and R3 are chosen independently from the group which consists of the alkyl and phenyl which may have branching of carbon numbers 1-5, a benzyl, and an allyl compound, respectively. As R2 and R3, specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, and an isopentyl are mentioned, and they are a methyl, ethyl, a phenyl, or a benzyl preferably.

[0045] R4 is an allyl compound (-CH2 CH=CH2), the alkyl which may have branching of carbon numbers 1-5, and [0046].

[Formula 17]



[0047] a shell -- it is one chosen from a group and R14-R19 are the alkyls which may have branching

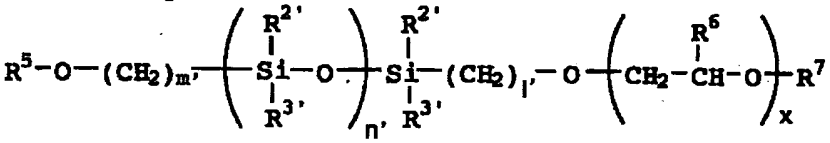
of a hydrogen atom or carbon numbers 1-5 As R4, specifically, -C(=O) C(CH3) =CH2, -C(=O) CH=CHCH3, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, and an isopentyl are mentioned, and they are a methyl and -C(=O) C(CH3) =CH2 preferably.

[0048] m is 1-5. n -- 1-20 -- it is 1-10 preferably l -- 1-20 -- it is 1-10 preferably When m, n, and l are this range, since the compound shown with a chemical formula 1 is excellent in solubility, reactivity, and the cell performance side, it is desirable.

[0049] As such a polysiloxane compound, -C(=O) C(CH3) =CH2 and R2 A methyl, [R1] The polysiloxane compound whose R4 R3 is a methyl and is a methyl, and m, n and l3, Ethyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 3 or whose 5l 2 or 3n is [R4] 3 or 5 for a methyl and m, A phenyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l a methyl and m are 3 or 5 for R4, A benzyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l a methyl and m are 3 or 5 for R4, A phenyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] R4 -- ethyl and m= -- 2 or 3, and n= -- 3 or 5, and l= -- the polysiloxane compound which is 3 or 5 -- A benzyl and R3 A methyl, [R1] [-C(=O) C(CH3) =CH2 and R2] The polysiloxane compound 2 or 3, and whose n and l ethyl and m are 3 or 5 for R4, Since it excels in ********, reactivity, and the cell performance side, it is used preferably, and the polysiloxane compound a methyl and whose R4 are [R1 / -C(=O) C(CH3) =CH2 and R2] a methyl, and m, n and l3 also especially in these for a methyl and R3 is desirable.

[0050] Next, a polysiloxane-polyoxyalkylene compound is explained. The compound concerned is chemical formula 2: [0051].

[Formula 18]

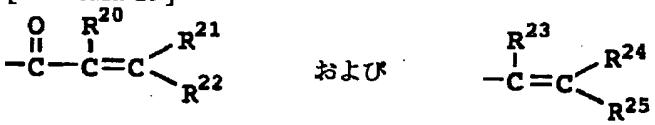


化学式2

[0052] It is come out and shown. R2' and R3' are chosen from the group which consists of the alkyl and phenyl which may have independently branching of carbon numbers 1-5, respectively, a benzyl, and an allyl compound among a formula. Specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned, and they are a methyl, ethyl, a phenyl, or a benzyl preferably.

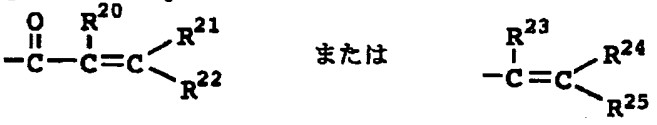
[0053] R5 and R7 are the alkyl which may have branching of carbon numbers 1-5, and [0054] independently, respectively.

[Formula 19]



[0055] a shell -- it chooses from a group -- having -- at least one [however,] of R5 and the R7 -- [0056]

[Formula 20]



[0057] It comes out, and it is and R20-R25 are the alkyls which may have independently branching of a hydrogen atom or carbon numbers 1-5, respectively. As R5 or R7, specifically, -C(=O) C(CH3) =CH2, -C(=O) CH=CHCH3, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned, and they are -C(=O) C(CH3) =CH2 or a methyl preferably.

[0058] R6 is the alkyl which may have branching of a hydrogen atom or carbon numbers 1-5, and, specifically, a methyl, ethyl, a propyl, an isopropyl, n-butyl, sec-butyl, ter-butyl, a pentyl, an isopentyl, etc. are mentioned as this alkyl. It is a hydrogen atom preferably as R6.

[0059] m' is 1-5. n' is 1-20 and is 1-10 preferably. l' is 1-20 and is 1-10 preferably. x is 1-15. m -- ' -- n -- ' -- l -- ' -- and -- x -- this -- the range -- it is -- the time -- a chemical formula -- two -- being shown -- having -- a compound -- solubility -- reactivity -- and -- a cell -- a performance -- a field -- excelling -- **** -- a sake -- being desirable .

[0060] As a polysiloxane-polyoxyalkylene compound shown with such a chemical formula 2 -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a methyl and R3'] [a methyl and R5] R7 A methyl, m', the polysiloxane-polyoxyalkylene compound whose x n' and l' are 3 and is 5, -C(=O) C (CH3) =CH2 and R6 A hydrogen atom, [R2'] [ethyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a benzyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2'] [a phenyl and R3'] [a methyl and R5] The polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m', -C(=O) C (CH3) =CH2 and R6 A hydrogen atom, [R2'] [a benzyl and R3'] [a methyl and R5] Since the polysiloxane-polyoxyalkylene compound 3, or whose 5 and x 2 or 3, n', and l' are [R7] 3, 5, or 10, respectively for a methyl and m' is excellent in solubility, reactivity, and the cell performance side, it is used preferably. these -- inside -- **** -- especially -- R -- two -- ' -- a methyl -- R -- three -- ' -- a methyl -- R -- five - C (=O) -- C (CH3) -- = -- CH -- two -- R -- six -- a hydrogen atom -- R -- seven -- a methyl -- m -- ' -- n -- ' -- and -- l -- ' -- three -- x -- five -- it is -- a polysiloxane -- a compound -- being desirable .

[0061] As for the content of the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with a chemical formula 1, it is desirable that it is 0.1 - 10 mass section on the basis of the constituent 100 mass section. The effect of a performance improvement of the cell according to addition of these compounds when a content is under the 0.1 mass section here is low, and when exceeding 10 mass sections, there is a possibility that the performance of a cell may fall.

[0062] Although the polysiloxane compound shown with a chemical formula 1 can make for example, a dihydroxy end polysiloxane and an acryloyl chloride able to react and can manufacture a polysiloxane compound, it is not limited to this but can be manufactured by the method of this business world common knowledge. Similarly, the polysiloxane-polyoxyalkylene compound shown with a chemical formula 2 can be manufactured by various methods. For example, although an acryloyl chloride can be made to be able to react to the polysiloxane-polyoxyalkylene which a dihydroxy end polysiloxane and ethylene oxide are made to react, and is manufactured and a polysiloxane-polyoxyalkylene compound can be manufactured, it is not limited to this but can manufacture by the method of this business world common knowledge.

[0063] Next, a polyethylene-glycol derivative is explained. The polyethylene-glycol derivative concerned is chemical formula 3: [0064].

[Formula 21]

$$H_2C=C(R')C(=O)(OCH_2CH_2)_zOR''$$

化学式3

[0065] It is come out and shown. R' is a hydrogen atom or CH3 among a formula, R" is a hydrogen atom, -C(=O) CH=CH2, or -C(=O) C(CH3) =CH2, and z is 1-20. Polyethylene glycol dimethacrylate, polyethylene-glycol diacrylate, polyethylene glycol monomethacrylate, polyethylene-glycol monoacrylate, etc. are mentioned preferably as such a polyethylene-glycol derivative. These may be used independently, respectively, and it may mix and they may be used. By using especially polyethylene glycol dimethacrylate, polyethylene glycol monomethacrylate, or those mixture, since the performance of a cell can be raised, it is desirable.

[0066] The range of the content of the polyethylene-glycol derivative concerned has desirable 0.4 - 50 mass section on the basis of the constituent 100 mass section. When a content is under the 0.4 mass section here, hardening reactivity falls, and when exceeding 50 mass sections, there is a possibility that the performance of a cell may fall. However, since the range of the content of a polyethylene-

glycol derivative is various by what is used, it is not this limitation. Hereafter, the range of a desirable content is described concretely. For example, as for the content of polyethylene glycol dimethacrylate, it is desirable that it is 0.5 - 50 mass section on the basis of the constituent 100 mass section, and, as for the content of polyethylene glycol monomethacrylate, it is desirable that it is 0.4 - 50 mass section on the basis of the constituent 100 mass section.
[0067] Next, the organic solvent containing the lithium salt of this invention is explained. It is desirable that it is or more [it is chosen from the group which consists of a lithium perchlorate (LiClO4), a 4 boron-fluoride lithium (LiBF4), a 6 fluoride / phosphorus / lithium (LiPF6), a 6 fluoride arsenic lithium (LiAsF6), a 3 fluoride methansulfonic acid lithium (LiCF3SO3), and a RICHIUMUBISU trifluoromethane sulfonyl amide (LiN2 (CF3SO2)) as lithium salt] one. It is LiPF6 especially preferably. Of course, well-known lithium salt is usable at the technical field concerned besides the above.
[0068] It is desirable that it is or more [it is chosen from the group which consists of propylene carbonate, ethylene carbonate, dimethyl carbonate, methylethyl carbonate, diethyl carbonate, BINIRUREN carbonate, triglyme, tetraglyme, and gamma-butyrolactone as an organic solvent] one. Especially, preferably, it is ethylene carbonate (EC), dimethyl carbonate (DMC), and dimethyl carbonate (DEC), and these may be used independently, and you may mix and use by the suitable volume ratio. Of course, a well-known organic solvent is usable at the technical field concerned besides the above.
[0069] As for the content of the organic solvent containing such lithium salt, it is desirable that it is 50 - 97 mass section on the basis of the constituent 100 mass section. And as for the content of lithium salt, it is desirable that it is 0.5-3 mols to organic-solvent 1L. When the content of an organic solvent and lithium salt separates from the aforementioned range here, there is a possibility that the performance of a cell may fall.
[0070] As for the constituent of this invention, it is still more desirable that ethoxylation trimethylolpropane triacrylate (ethoxylated trimethylol propane triacrylate) is included. The hardening reaction of a constituent can be promoted by including such a compound. The content of ethoxylation trimethylolpropane triacrylate has a possibility that the performance of a cell may fall, when it is desirable that it is 0 - 5 mass section and it exceeds 5 mass sections on the basis of the constituent 100 mass section here.
[0071] The constituent for forming the macromolecule gel electrolyte of this invention can be obtained by mixing the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with the chemical formula 1 described above, a polyethylene-glycol derivative, and the organic solvent containing lithium salt.
[0072] Furthermore, as for the aforementioned constituent, it is desirable that one or more polymerization initiators chosen from a benzophenone, a benzoyl peroxide, an acetyl peroxide, a lauroyl peroxide, and the group that consists of an azobisisobutyronitril are included, and, generally especially an azobisisobutyronitril is used. Generally, on the basis of the constituent 100 mass section, although the content of a polymerization initiator is 0.1 - 5 mass section, it should choose each suitable content according to the kind here.
[0073] The macromolecule gel electrolyte of this invention is manufactured by hardening the constituent containing an above-mentioned component. Hardening is desirable and thermal polymerization, the polymerization by the electron beam, or the polymerization by UV is used. Each polymerization method is explained in detail below.
[0074] Thermal polymerization is performed by processing a constituent in predetermined polymerization time in the oven adjusted by polymerization temperature. In the case of less than 60 degrees C, as for polymerization temperature, it is desirable that it is 60-100 degrees C, and there is a possibility that a polymerization may become inadequate, and if 100 degrees C is exceeded on the other hand, an expansion phenomenon may produce it here. carrying out the polymerization of the polymerization time for example, within a case -- or although it is not limited since it is required to adjust suitably by whether a polymerization is carried out in the state where it cast for the film, generally it is about 4 hours
[0075] The polymerization by the electron beam is made when predetermined carries out time

irradiation of the electron beam at a constituent. The output of this electron beam is usually 1.5eV or more, and in the case of less than 1.5eV, the long setting time is needed or it has fear, like hardening becomes inadequate here. Generally, as for time to irradiate an electron beam, it is desirable that it is 30 seconds or more, and when time is less than 30 seconds, it has a possibility that a polymerization may become inadequate. The polymerization by such electron beam can shorten reaction time compared with above-mentioned thermal polymerization, and has the advantage of not needing a polymerization initiator further.

[0076] The polymerization by UV is made when predetermined carries out time irradiation of the UV at a constituent. Generally, the wavelength of this UV has a possibility that a polymerization may become inadequate, when it is desirable that it is 250-365nm and it separates from this range here. Generally, as for time to irradiate UV, it is desirable that it is 30 seconds or more, and when time is less than 30 seconds, it has a possibility that a polymerization may become inadequate. The polymerization by such UV has the advantage that reaction time can be shortened compared with above-mentioned thermal polymerization.

[0077] Next, how to manufacture a macromolecule gel electrolyte using the constituent mentioned above is explained. It prepares within the limits of the content which mentioned each component above first. These contents should be chosen the optimal in consideration of the performance of the electrochemical stability of the lithium cell manufactured, and a cell etc. Next, the polysiloxane-polyoxyalkylene compound shown with the polysiloxane compound or chemical formula 2 shown with a chemical formula 1, the polyethylene-glycol derivative shown with a chemical formula 3, and ethoxylation trimethylolpropane triacrylate are mixed. Into this mixture, the above-mentioned polymerization initiator is added further if needed, the organic solvent which contains lithium salt further is added and stirred into it, and a uniform constituent is obtained. The obtained constituent is hardened by the thermal polymerization mentioned above, the polymerization by the electron beam, or the polymerization by UV.

[0078] Next, the lithium cell using the macromolecule gel electrolyte of this invention is explained. The lithium cell of this invention possesses the case having the electrode assembly containing the separator which intervenes between a cathode, an anode, and the aforementioned cathode and the aforementioned anode, the macromolecule gel electrolyte of this invention mentioned above, and the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte.

[0079] Explanation of the cathode active material constituent and anode active material constituent which are first used as a cathode or an anode uses preferably for a cathode the cathode active material constituent which generally consists of lithium content oxides, such as LiCoO2. The anode active material constituent which generally contains carbon, graphite, etc. is preferably used for an anode, and, generally a mezzo-soprano carbon fiber is desirable especially as carbon.

[0080] Furthermore, these active material constituent may contain the electric conduction agent, the binder, and the solvent suitably. As an electric conduction agent, although not limited to these, carbon black etc. is used preferably. Since the conductivity of an electrode active material layer falls when it is desirable that it is 1 - 20 mass section and a content exceeds 20 mass sections here on the basis of the electrode active material (for example, LiCoO2) 100 mass section, and the content of an electrode active material becomes low relatively in being under 1 mass section, the content of an electric conduction agent is not desirable here.

[0081] As a binder, a vinylidene fluoride-hexafluoropropylene copolymer (VdF/HFP copolymer), poly vinylidene fluoride, a polyacrylonitrile, the poly methyl methacrylic rate, or its mixture is used preferably, and, generally especially poly vinylidene fluoride is desirable. As for the content of a binder, it is desirable that it is 2 - 30 mass section on the basis of the electrode active material 100 mass section. When a content is the aforementioned range here, it excels in the bonding strength of the electrode active material to an electrode charge collector.

[0082] As a solvent, if generally used for a lithium cell, all are usable, for example, an acetone and N-methyl pyrrolidone are mentioned, and it combines with other components, and is chosen suitably.

[0083] Although separator will not be restricted especially if used in the field concerned, it is desirable that it is the sheet which combined a polyethylene sheet, a polypropylene sheet, or these. It is preferably used by the reason especially for being easy to roll round polyethylene separator, and

polypropylene / polyethylene / three layer separator of polypropylene. Specifically, the polyethylene separator by the morning-sun chemical-industry company is used.

[0084] Although all are usable if it is the type used in this industry, since especially a pouch type can prevent external disclosure of a gel electrolyte good, the case having the aforementioned electrode assembly and the aforementioned macromolecule gel electrolyte containing such a cathode, an anode, and separator is desirable.

[0085] Next, the manufacture method of the lithium cell of this invention is explained. First, an electrode active material layer is formed on a charge collector using an above-mentioned anode active material constituent or an above-mentioned cathode active material constituent, and a cathode electrode board and an anode electrode board are produced. An electrode active material layer is formed here by the method of coating each active material constituent directly on a charge collector, and the method of carrying out the lamination of the film of the active material constituent obtained from this base material by exfoliating after coating and drying each active material constituent in the base material upper part separately with a doctor blade on a charge collector. If an active material layer can be supported, such a base material can use all, for example, polyethylene terephthal rate films, such as a Mylar film (Du Pont make), are suitable for it.

[0086] Next, separator is inserted between the cathode electrode boards and anode electrode boards which were obtained, and an electrode assembly is formed in it. Even if it forms the assembly concerned by rolling up using the jelly roll method (drawing 1), you may form it in the Bayh cellular structure here (drawing 2). Thus, the obtained assembly is installed in a case.

[0087] Next, the macromolecule gel electrolyte of this invention is formed by preparing the constituent for forming the macromolecule gel electrolyte of this invention in a case, and hardening it. The method of hardening the case which poured in the constituent for forming the macromolecule gel electrolyte of this invention into the case as one method, sealed, and was acquired by the polymerization by the electron beam mentioned above or the polymerization by UV is mentioned. Thus, when carrying out the polymerization of the constituent within a case, the effect by thermal polymerization can stiffen a constituent most efficiently.

[0088] After casting the constituent for forming the electrolyte of this invention in either an anode electrode board or a cathode electrode board and the front face of both these with a doctor blade as other methods, the method of hardening by the polymerization by the electron beam mentioned above or the polymerization by UV is also desirable. Thus, the lithium cell of this invention can be obtained.

[0089] The lithium cell of this invention is applicable to all of lithium rechargeable batteries, such as a lithium primary cell, a lithium ion polymer battery, or a lithium ion battery.

[Translation done.]

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EXAMPLE

[Example] Hereafter, an example is given and this invention is explained more to a detail.
[0091] Added poly vinylidene fluoride 15g to <production of macromolecule gel electrolyte and lithium cell using it> example 1 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and acetylene black (tradename : super - P, product made from MMM) 15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was produced.
[0092] The aforementioned cathode active material constituent was coated using the doctor blade which has the gap of 320 micrometers on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, it dried and the unit cathode electrode board was produced.
[0093] On the other hand, poly vinylidene fluoride 50g was added to acetone 600ml, mix for 2 hours, it was made to dissolve with a ball mill, mixture was obtained, mezzo-soprano carbon fiber 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was produced.
[0094] The aforementioned anode active material constituent was coated using the doctor blade which has the gap of 420 micrometers on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, it dried and the unit anode electrode board was produced.
[0095] Polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm] and a thickness of 18 micrometers was made to intervene between the aforementioned cathode electrode board and the aforementioned anode electrode board, this was rolled round by the jelly roll method, and the electrode assembly was produced. This electrode assembly was put into the pouch.
[0096] On the other hand, it sets in a chemical formula 1. a methyl and R1 -C(=O) C(CH3) =CH 2 and m, [R4 R2, and R3] 0.2g [of polysiloxane compounds], and polyethylene-glycol JIMETA krill rate 1.8g whose l is n and3, Polyethylene-glycol monochrome methacrylic rate 0.5g, ethoxylation trimethylolpropane triacrylate 0.5g, Azobisisobutyronitril 0.1g, and (ethylene carbonate EC) / dimethyl carbonate (DMC) / dimethyl carbonate (DEC) = 30g of organic solvents which are 3:3:1 (volume ratio) and contain LiPF6 1M is mixed. The constituent for forming a macromolecule gel electrolyte was prepared. 3g of this constituent was poured into the pouch containing the electrode assembly obtained by the above-mentioned method, and it was sealed. The lithium cell of this invention was completed by as a result processing an object in the oven adjusted at 80 degrees C for 4 hours.
[0097] Added poly vinylidene fluoride 15g to example 2 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. After adding LiCoO2470g and super-P (product made from MMM)15g into this mixture, this was mixed for 5 hours and the cathode active material constituent was produced.
[0098] The doctor blade which has the gap of 320 micrometers for the obtained cathode active material constituent was used, and it coated on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, and it dried and the unit cathode electrode board was produced.
[0099] Then, 0.2g [of the same polysiloxane compounds as an example 1] and polyethylene-glycol JIMETA krill rate 1.8g, polyethylene-glycol monochrome methacrylic rate 0.5g, ethoxylation trimethylolpropane triacrylate 0.5g, benzophenone 0.1g, and 30g of organic solvents which are

EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M were mixed, and the constituent for forming a macromolecule gel electrolyte was manufactured. After using and casting a doctor blade for the cathode electrode board which manufactured this constituent above, 365nm UV is irradiated for 1.5 hours, and was stiffened, and the cathode electrode in which the macromolecule gel electrolyte was formed was obtained.

[0100] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and the constituent was obtained. Mezzo-soprano carbon fiber 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0101] The doctor blade which has the gap of 420 micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0102] After making polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm], and a thickness of 18 micrometers intervene between the aforementioned cathode electrode boards and the aforementioned anode electrode boards with which the macromolecule gel electrolyte was formed, this was rolled round by the jelly roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch and the lithium cell was completed.

[0103] If it removed having made the anode electrode board cast and harden the constituent for forming an example 3 macromolecule gel electrolyte instead of a cathode electrode board, the lithium cell was produced by the same method as an example 2.

[0104] If it removed having cast the constituent for forming an example 4 macromolecule gel electrolyte for both the cathode electrode board and the anode electrode board, the lithium cell was produced by the same method as an example 2.

[0105] Added poly vinylidene fluoride 15g to example 5 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and super-P (product made from MMM)15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was prepared.

[0106] The doctor blade which has the gap of 320 micrometers for the aforementioned cathode active material constituent was used, and it coated on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, and it dried and the unit cathode electrode board was produced.

[0107] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. Mezzo-soprano carbon fiber (MCF) 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0108] The doctor blade which has the gap of 420 micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0109] After making polyethylene separator (morning-sun chemical-industry company) with 18 micrometers [in thickness], and a width of face of 5.25cm intervene between the aforementioned cathode electrode board and the aforementioned anode electrode board, this was rolled round by the ZERU roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch.

[0110] On the other hand, it sets in a chemical formula 2. -C(=O) C(CH3) =CH2 and R6 A hydrogen atom, [R2', R3', and R7] [a methyl and R5] 3 and x 0.2g of 5 polysiloxane-polyoxyalkylene compounds, [m', n', and l'] Polyethylene-glycol JIMETAKURIRURE - TO1.8g, polyethylene-glycol monochrome methacrylic rate 1g, Ethoxylation trimethylolpropane triacrylate 0.05g, azobisisobutyronitril 0.01g, and 30g of organic solvents which are EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M are mixed. The constituent for forming a macromolecule gel electrolyte was manufactured. 3g of this constituent was poured into the pouch cell obtained in the

example 1, and this was sealed. Then, the lithium cell was produced by processing join fruit in the oven adjusted at 80 degrees C for 4 hours.

[0111] Added poly vinylidene fluoride 15g to example 6 acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. LiCoO2470g and super-P (product made from MMM)15g were added into this mixture, it mixed for 5 hours, and the cathode active material constituent was prepared.

[0112] The aforementioned cathode active material constituent was coated using the doctor blade which has the gap of 320 micrometers on the aluminum thin film with 147 micrometers [in thickness], and a width of face of 4.9cm, it dried and the unit cathode electrode board was produced.

[0113] Then, 0.2g [of the same polysiloxane-polyoxyalkylene compounds as an example 5] and polyethylene-glycol JIMETA krill rate 1.8g, polyethylene-glycol monochrome methacrylic rate 1g, ethoxylation trimethylolpropane triacrylate 0.05g, benzophenone 0.01g, and 30g of organic solvents which are EC:DMC:DEC=3:3:1 (volume ratio) and contain LiPF6 by 1M were mixed, and the constituent for forming a macromolecule gel electrolyte was prepared. After using and casting a doctor blade for the cathode electrode board which manufactured this constituent above, UV with a wavelength of 365nm was irradiated for 1.5 hours, and was stiffened.

[0114] On the other hand, added poly vinylidene fluoride 50g to acetone 600ml, and mix for 2 hours, it was made to dissolve with a ball mill, and mixture was obtained. Mezzo-soprano carbon fiber (MCF) 449g and 1g of oxalic acid were added into this mixture, it mixed for 5 hours, and the anode active material constituent was prepared.

[0115] The doctor blade which has the gap of 420.micrometers for the aforementioned anode active material constituent was used, and it coated on the copper thin film with 178 micrometers [in thickness], and a width of face of 5.1cm, and it dried and the unit anode electrode board was produced.

[0116] After making polyethylene separator (morning-sun chemical-industry company) with a width of face [of 5.25cm], and a thickness of 18 micrometers intervene between the cathode electrode boards and anode electrode boards with which the macromolecule gel electrolyte was formed, this was rolled round by the jelly roll method and the electrode assembly was produced. This electrode assembly was put in in the pouch and the lithium cell was produced.

[0117] If it removed having cast the constituent for forming an example 7 macromolecule gel electrolyte for the anode electrode board instead of the cathode electrode board, the lithium cell was produced by the same method as an example 6.

[0118] If it removed having cast the constituent for forming an example 8 macromolecule gel electrolyte for both the cathode electrode board and the anode electrode board, the lithium cell was produced by the same method as an example 6.

[0119] Instead of the constituent for forming the macromolecule gel electrolyte of the example this invention of comparison, it is 1M. If it removed having used the mixed solution (Ube Industries, Ltd. make) which contains LiPF6 and EC/DMC/DEC by the volume ratio 3:3:4, the lithium cell was produced by the same method as an example 1.

[0120] The lithium cell of the <characterization of cell> test-method examples 1-8 and the example of comparison evaluated reliability and safety by the life characteristic test, the penetration examination, the expansion test at the time of elevated-temperature neglect (85 degrees C), and the liquid spill examination under pressure 40 kgf/cm2.

[0121] The life characteristic test was made by observing the volume change of the cell according charge and discharge to a 100 cycle deed and it using the lithium cell of examples 1-8 and the example 1 of comparison. This evaluation made it desirable for there to be few volume changes.

[0122] The penetration experiment made the nail with a diameter of 5mm penetrate in the direction perpendicular to the major axis of after 3-hour charge and a cell in the center with the current of 0.2C, and investigated the existence of the ignition phenomenon of a cell, and a rupture phenomenon.

[0123] After leaving an expansion test at 85 degrees C after 3-hour charge in the current of 0.2C for 4 hours, it measured and evaluated the thickness of a cell. The thickness of the cell after neglect presupposed that it is good, when initial thickness became 110% or less.

[0124] The disclosure examination investigated the existence of a liquid spill, after pressurizing the

cell for 10 seconds by the pressure of 40 kgf/cm2.

[0125] Test-result drawing 3 is a graph which shows the life property of the lithium cell of an example 1 and the example of comparison. According to this, the volume decrease of the cell of an example 1 was almost of the same grade as the example of comparison. Moreover, in the penetration examination, neither ignition nor rupture takes place, but is maintaining 110% of the initial thickness of a cell also in the expansion test further, and did not carry out a liquid spill from inside in the disclosure examination further. Since examples 2-8 also showed the almost same result, they were found by that the lithium cell of this invention has the outstanding life property.

[0126] As mentioned above, since the lithium cell of examples 1-8 can maintain the gel excellent in the electrolytic solution, it turns out that expansion of external disclosure of the electrolytic solution, the electrode assembly by the electrolytic solution, or a pouch can be suppressed, and it has the reliability and the safety superior to the cell of the example of comparison.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] It is the decomposition perspective diagram having shown typically an example of the lithium ion battery which used the general pouch.
[Drawing 2] It is the decomposition perspective diagram having shown an example of the conventional lithium ion polymer battery typically.
[Drawing 3] It is the graph which shows the result which measured the life property of the lithium cell of an example 1 and the example of comparison.
[Description of Notations]
10 21 Electrode assembly
11 Cathode
12 Anode
13 Separator
14 22 Case
15 23 Cathode tap
15', 23' Anode tap
16, 16', 24, 24' Electrode terminal (or lead wire)

[Translation done.]

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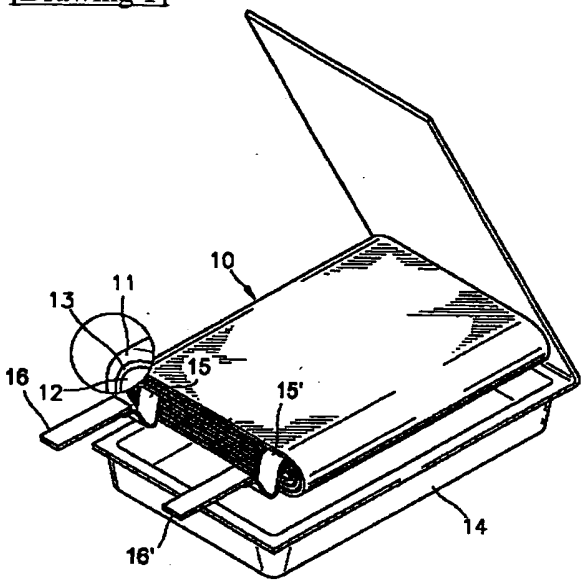
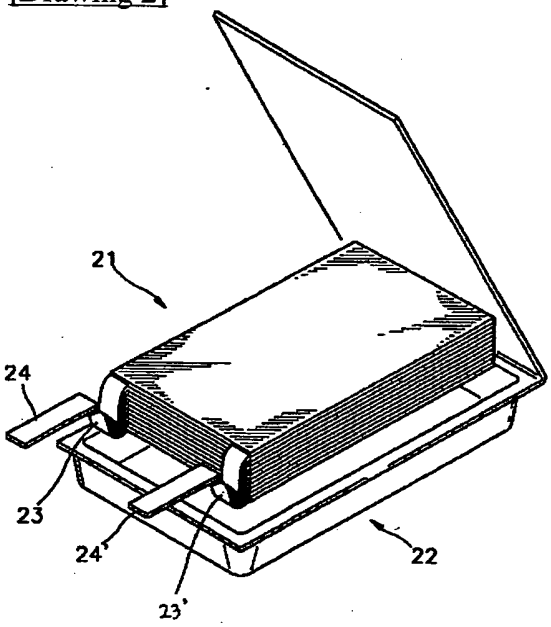
DRAWINGS

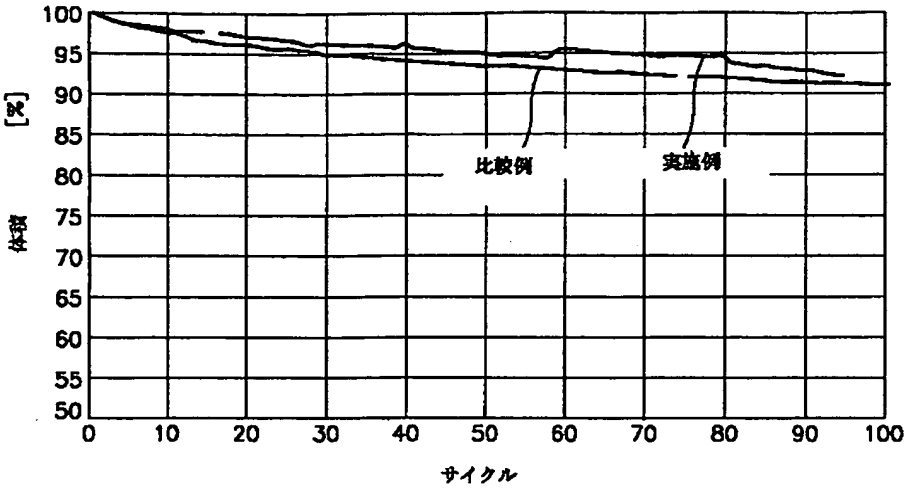
[Drawing 1]



[Drawing 2]



[Drawing 3]



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